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NUCLEAR WASTE

**COMMENTARY ON THE STATE OF KNOWLEDGE OF THE
ORIGINS OF THE YUCCA MOUNTAIN CALCITE VEINS**

SPECIAL REPORT No.17
CONTRACT No. 94/96.0003

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SPECIAL REPORT Submitted to the
Nuclear Waste Project Office
State of Nevada

August, 1994

Authored by:

Dr. Charles Archambeau

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COMMENTARY ON THE STATE OF KNOWLEDGE OF THE ORIGINS OF THE YUCCA MOUNTAIN CALCITE VEINS

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August 12, 1994

Martin Steindler
Chairman, Advisory Committee on Nuclear Waste
Nuclear Regulatory Commission
7920 Norfolk Ave.
Bethesda, Maryland

Dear Mr. Steindler:

Technology and Resource Assessment Corporation-North America (TRAC-NA) has a contract with the State of Nevada to investigate the geologic and hydrologic stability of the proposed Yucca Mountain Repository site. In the course of our work we have also tried to keep current on the activities of your committee. In this regard we recently reviewed the proceedings of your 64th ACNN meeting and felt we should comment on one of the studies discussed during the meeting, in particular the study by James Wood as presented by William Ott of the NRC.

We have done similar studies involving Carbon and Oxygen isotopes as well as fluid inclusion studies at, and in the vicinity of, Yucca Mountain. In addition we have, along with our consultants, studied the petrology and mineralogy of the rocks at the site and investigated the occurrence of Uranium, Lead and Strontium isotopes in the abundant calcite-opal veins at the surface, and at depth, at Yucca Mountain.

We would agree that the Oxygen and Carbon isotopic signatures for the calcite-opal veins are inconclusive, by themselves, as to the origins of the veins. However, we note that when homogenization temperature data from inclusions are obtained and considered, then the combined evidence becomes more conclusive. If the complete data set involving the other isotopes and the mineralogy and chemistry of the altered tuffs is also taken into account, then we conclude that there can be little doubt as to the origins of the veins and calcretes at Yucca Mountain; they are epithermal, that is generally moderate temperature deposits from hypogene (up-welling) fluids. Consequently, these are "thermogenic" deposits, as are those described by Wood.

A more detailed description of our conclusions is enclosed in a Nevada State report: "Past Research and Results and Plans for FY '95", presented to the Nuclear Waste Projects Office, State of Nevada, July 1994. This document, while brief and without much technical detail, does summarize more of the basis for our conclusions. Should you want to pursue the complete and detailed technical basis for these conclusions, we can send you appropriate technical reports that have been submitted to the State of Nevada. However, we have enclosed a paper by Hill et. al. that has been submitted to *Science* for publication that does cover, in a condensed format, some of the essential technical background and data. The conclusions by these authors are similar to ours.

We have noted in your proceedings a suggestion of the rather common perception that (in the words of Mr. Hatcher on p. 51 of the proceedings): "I thought a lot of the evidence from Yucca Mountain indicated these things were meteoric in origin and not hydrothermal or thermogenic, as you say here." To which Mr. Ott replied: "Right, that is correct." These are not uncommon perceptions and they are fostered by private and public statements, government reports and (even) reviewed papers in the scientific literature by Yucca Mountain project investigations that conclude that the surficial calcite-opal veins and calcretes are of pedogenic origin (meaning, in this context, that they are derived from wind blown dust and deposited by rain water) and even that the calcite-opal veins found at great depth (as found in drill core samples) are of the same origin. The common flaw in all of these statements, written or oral, is that the authors start out with assumptions (usually unstated) that are not justified and then proceed to use equivocal or incomplete data as support for their conclusions while ignoring other data (without any stated justification) that does not appear to be compatible. In some cases authors ignore some of their own data (that is not consistent with the conclusion they wish to draw) and, in other cases, authors have actually misrepresented the data and conclusions of other researchers to formulate a desired conclusion.

Examples of published conclusions based on equivocal data that have led to the perception that the field evidence supports a pedogenic origin for the young (ages less than 10 Ma) calcite-opal veins at Yucca Mountain are the papers by Quade and Cerling (*Science*, 1990) and Stuckless et al. (*Science*, 1991).

In this regard Quade and Cerling consider Carbon and Oxygen isotopic data from the calcites at Trench 14 on Yucca Mountain and compare the observations to "Holocene pedogenic calcite deposits" in the area. They find, from the latter, a trend of decreasing

$\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values (increasingly more negative PDB values) with altitude for the "pedogenic" samples. (The $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values are normalized ratios of ^{13}C to ^{12}C and ^{18}O to ^{16}O .) They also find that the $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values for samples at Trench 14 have a narrow range that are much *lower*, for both $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$, than the values for the "pedogenic" samples at the same altitude as Trench 14, but that are in the same range as for "pedogenic" samples obtained at an altitude some 750 to 790 meters *higher* than Trench 14. They then argue that this indicates that the Trench 14 carbonates must have been deposited by rainwater during pluvial ice ages when the climate was cooler by an amount roughly corresponding to the mean temperature difference between the current averages at the Trench 14 elevation and that at 750 meters higher. In doing so they note that the $\delta^{18}\text{O}$ ground water values during cool ice ages are known to be less than that presently observed by an amount that is roughly that required to bring the observed Trench 14 values into agreement with the Holocene (less than 10,000 years old) "pedogenic" samples at the same altitude. However, they do not state how much and in what direction (increase or decrease) the $\delta^{13}\text{C}$ would change with a cooler climate, leaving the reader to infer (based on previous statements) that the vegetation would change to become similar to that now at the higher altitude and that this change would result in lower (more negative) values for $\delta^{13}\text{C}$ by the amount required. Further, they imply that this would be due to a difference in the new plant fractionization of the Carbon that favors more negative values in $\delta^{13}\text{C}$. The authors then conclude that the Trench 14 carbonates are consistent with a pedogenic origin and that the deposition of the calcite-opal veins at Trench 14 occurred during several ice ages over a span of about 300,000 years.

There are several problems with both their line of reasoning and their conclusions. First is the fact that changes in $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ due to climate changes are observed to be *anti-correlated*; that is at the nearby Devils Hole, *increases* in the $\delta^{18}\text{O}$ value in the spring carbonates due to *warming* are found to be clearly accompanied by significant *decreases* in $\delta^{13}\text{C}$ (Coplen et al., *Science* v.263, 1994). Conversely, during *cooling* periods the $\delta^{18}\text{O}$ *decreases* significantly and the $\delta^{13}\text{C}$ increases slightly or does not change at all. This is in apparent disagreement with the change due to a cooler climate inferred by Quade and Cerling, in that the large *decrease* in $\delta^{13}\text{C}$ inferred by them is not seen in the Devils Hole calcite and in fact a slight increase occurs. Since the cool climate change in vegetation that is inferred to cause the required decrease in the $\delta^{13}\text{C}$ value for a pedogenic calcite at the Trench 14 altitude should also cause a regional change in the vegetation and a decrease in the $\delta^{13}\text{C}$ value in the ground water and in

the carbonates deposited from such waters at Devils Hole, there is clearly reason to question the Quade and Cerling inference. Furthermore, warm climatic periods clearly *decrease* the $\delta^{13}\text{C}$ value by a large amount (while increasing the $\delta^{18}\text{O}$ value) and, as noted by (Coplen et al., *Science* v.263, 1994), the $\delta^{13}\text{C}$ change would be expected to be controlled by an accompanying change in vegetation (type and density). However, the $\delta^{13}\text{C}$ change during a warming period is again in the opposite sense (a decrease) from that to be expected from the Quade and Cerling argument, where the latter would imply an *increase* in the $\delta^{13}\text{C}$ value during a warm period due to a change in the type of vegetation. These major discrepancies are not addressed by Quade and Cerling and cannot be dismissed. On these grounds alone there is no logical basis for their conclusion of a pedogenic origin for the Trench 14 calcite veins.

Other difficulties with Quade and Cerling's analysis arise when more of the available data are used to evaluate the likelihood of a pedogenic origin of the calcite veins at Trench 14. In particular, in addition to Devils Hole calcites, calcites at other spring deposits in the vicinity of Yucca Mountain (at the southern Crater Flats spring deposit and the Whamonic Springs deposit) show that these springs, taken as a group, produce calcites having a wide range of $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ values, with the values observed at Trench 14 occurring in the middle of this range and with the "pedogenic" values given by Quade and Cerling near one end of the range, but well within it. Therefore, while the Devils Hole calcites have different $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ values than those at Trench 14, they are also even more different than those at some of the other known spring deposits in the area. There is, therefore, no basis for rejecting the idea that Trench 14 calcites could be spring deposits simply because the Carbon and Oxygen isotopic ratios aren't the same as those at Devils Hole. There is, in fact, several physical reasons (temperature differences, different source rocks at depth) for expecting differences as well as the empirical evidence that large differences, are observed between known spring deposits. On the other hand, as noted earlier, an argument can be made for rejecting a pedogenic origin for the Trench 14 calcites, based on the observed O and C isotopic signature variations with climate.

In addition, beyond the incompatibilities with other data not addressed by Quade and Cerling, the implications of the conclusion reached strains credibility and common sense. That is, they have concluded that the Trench 14 calcites were deposited during ice ages over a span of at least 300,000 years. They are forced to the conclusion that deposition occurred essentially *only* during ice ages because the ranges of $\delta^{13}\text{C}$ and

$\delta^{18}\text{O}$ values found at Trench 14 are so narrow. (That is, if there had been significant pedogenic deposition at Trench 14 during warm periods, as well as during cold periods, there would be a very wide range of variation in both $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values observed as can be verified from the Devils Hole variations recorded over a period of 500,000 years. Since wide variations are not observed, this requires deposition over a narrow temperature range and at cool temperatures because of the low mean $\delta^{18}\text{O}$ value required for the rainwater.) To produce the massive and quite pure vertically banded calcite and opal veins observed by such an episodic pedogenic process requires that surface fractures, to considerable depth, would have to remain open and not be filled with debris between ice ages, or else fractures in one place (at Trench 14) would have to recurrently open just in time for each ice age! Added to this, there would have to occur a very rapid pedogenic deposition in a cool climate to maintain the required purity of the deposit and to accumulate the necessary volumes of calcite and opal. These are certainly very unlikely scenarios, and it is much more likely that a rapid depositional mechanism associated with faulting and involving ground water upwelling is responsible for these deposits.

Finally, it should be noted that Quade and Cerling implicitly assume that the "pedogenic" samples they use are derived from the evaporation of rain water that has acquired calcite from wind blown dust. However, it is as likely, if not more likely, that these deposits could be derived from secondary reworking of epithermal calcite deposits by rain water at and near Yucca Mountain. In this case some of the isotopic characteristics of the derived pedogenic deposits would reflect an epithermal origin and some not. In general for a reworked calcite, a Carbon 14 date would reflect the (average) age of pedogenic depositions and the $\delta^{18}\text{O}$ and $\delta^{14}\text{C}$ values would reflect the rain water isotopes, but the Strontium isotopic signature, for example, would be more characteristic of the parent epithermal deposit. In any case, the important point is that by simply asserting that the samples are "pedogenic", as do Quade and Cerling, does not define the real source of the calcite unless more than just Carbon and Oxygen isotopes are used. Since the real issue is: where did the calcite come from; is it derived from wind blown dust (and deposited through the evaporation of rain water) or is it from the underlying paleozoic and precambrian rocks (and deposited by upwelling ground water); it must (logically) be concluded that the comparative approach used by Quade and Cerling is incapable of resolving this critical issue. That is, such pedogenic deposits could derive their calcium from either existing surficial epithermal veins and calcretes deposited by upwelling ground water or from wind blown dust and one couldn't

distinguish which of these sources was involved on the basis of the Carbon and Oxygen isotopes alone. Therefore, not only is Quade and Cerling's argument for a pedogenic origin of the Trench 14 calcites flawed, but the presumption that correspondence with a pedogenic signature would mean that the Trench 14 calcium source is from wind blown dust, picked up and deposited by rain water, is also incorrect because a pedogenic signature in Carbon and Oxygen isotopes is not unique in this respect. (Furthermore, as noted earlier, some known spring deposits in the area have Carbon and Oxygen isotopic signatures that are the same as those for pedogenic deposits cited by Quade and Cerling, at the same altitude. This shows that the Carbon and Oxygen isotopes lack uniqueness to the degree that even if there is a match, one cannot conclude that an unknown calcite deposit is pedogenic; it could be either a spring or a rain water deposit.) In summary, it appears to us that Quade and Cerling's investigation, when evaluated with more of the pertinent data, indicates that a pedogenic origin for the Trench 14 calcite-opal veins is very unlikely, rather than indicating that it is likely or certain.

The paper by Stuckless et al. also suffers from the use of equivocal data in drawing unequivocal conclusions. In doing so the authors ignore pertinent information and data that would have precluded their conclusions. In this regard they quote Quade and Cerling's study of the Oxygen and Carbon isotopes as support for a pedogenic origin for the Trench 14 calcite and opal veins but do not, themselves, directly consider the O and C isotopic data in combination with the Strontium and Uranium isotopic data they consider. They simply assert that the O, C, Sr and U isotopic signatures are all compatible with a pedogenic origin based on their analysis of Sr and U data and Quade and Cerling's analysis of O and C data. As outlined above, it is evident that there are major difficulties with the Oxygen and Carbon isotopic argument advanced by Quade and Cerling when a more complete data set is used. Indeed, the O and C isotopic signatures for calcites at Trench 14 are, most likely, not compatible with pedogenic calcites in the area.

Aside from this, the arguments advanced by Stuckless et al. in their interpretation of the Uranium and Strontium isotopic data do not support an unambiguous interpretation of the Trench 14 calcites, nor of other similar deposits at Yucca Mountain. In this regard, these authors report the high values of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios measured for calcites in the Trench 14 veins. These values are much higher than those measured in the limestones that could supply the calcite in the dust for pedogenic deposition of these veins (.7125 on average for the veins compared to .7090 for the limestone), but are very near the

values observed at springs (eg. Ash Meadows, Devils Hole, Southern Crater Flats) in the vicinity of Yucca Mountain. The authors, however, assert that these high values were produced by a combination of the (low) values observed in wind blown dust plus contributions from the wall rock in the vicinity of the veins, which have high values. Of course it might be hypothesized that this mixing could have happened, but it is hardly a proof that it did, or even that it is likely.

Instead, it seems much more reasonable that the agreement of the Strontium signature at Trench 14 with observations at other springs, at various locations near Yucca Mountain, indicates a much greater likelihood that the deposits are from up-welling ground water with a high Strontium isotope ratio derived, in part, from deep seated paleozoic and precambrian rocks. In this case much of the alteration of the wall rock near calcite veins would be expected to be a consequence of the moderate to high temperature water intrusion which would also produce the high Strontium isotopic ratios in the altered tuffs. Therefore, upwelling water from considerable depth could produce the high Sr isotope ratios observed in both the calcites and in much of the wall rock and also be in agreement with observations from other spring deposited calcites in the area.

While Stuckless et al. note that the Trench 14 Strontium isotopic signature is the same as that of the water at springs in Ash Meadows, they dismiss this agreement as fortuitous, explaining the Ash Meadows value as being a mixture waters from different source areas that just happens to give nearly the same value as that at Trench 14. However such an explanation has elements of arbitrariness and special pleading, and certainly doesn't make sense when it is recognized that many known spring deposits at different locations and at different elevations in the Trench 14 vicinity have the same Strontium ratio signature. In this case it becomes much more difficult to advance a plausible argument that the agreement with many spring deposits is fortuitous and particularly so if the argument is of the type given by Stuckless, et al., since it would require water mixing from many different source areas, all magically giving the same, or nearly the same, Strontium isotopic ratio.

To make matters worse, Stuckless, et al. then state that "pedogenic" carbonate samples obtained by them, with many of these from Crater Flat, have Strontium ratios that are indistinguishable from the (Trench 14) vein carbonates. Given that there are many known spring carbonates having the same Strontium ratios as the vein carbonates as well, then this would say that most spring carbonates, vein carbonates and pedogenic

carbonates *all* have the same, or nearly the same, Strontium ratios. If this were true there would be no way to tell them apart on the basis of Strontium isotopes and no unambiguous way of determining the origin of an unknown deposit. But this is not the case, most pedogenic carbonates have lower Strontium ratios as noted, for example, by Marshall and Mahan (Radioactive Waste Management Conference, 1994). What appears to have happened is that Stuckless et al. have misidentified Crater Flat samples as pedogenic when they are actually from the Southern Crater Flats spring deposits and/or selected pedogenic samples derived from reworking of spring or vein deposits. In this latter case, depending on the degree of contamination during reworking, the Strontium ratio of the pedogenic deposit would be influenced by the ratio of the parent spring or vein calcite and have a high value, often comparable to a spring calcite. In cases when a pedogenic carbonate is less likely to be produced by reworking of a nearby spring or vein carbonate, the Strontium isotopic ratio value is typically that given by Marshall and Mahan, that is near .7116, and significantly lower than the .7125 average for spring and Trench 14 vein carbonates.

Therefore, the Stuckless et al. conclusion that Trench 14 vein carbonate Strontium ratios are essentially the same as pedogenic carbonate Strontium ratios is not correct. As a consequence it is also not correct to consider these vein carbonates as being compatible with ordinary pedogenic carbonates, either in this respect or with respect to Carbon and Oxygen isotopic characteristics.

One of the other main points of the Stuckless et al. paper was contained in the observation that the Strontium ratios of the Trench 14 carbonates are significantly higher than the ratios for the water in the Tertiary/Quaternary aquifer at Yucca Mountain and that this discordance precludes a genetic relationship between the ground water and the hydrogenic deposits. However this conclusion assumes that only this relatively shallow (and younger) water would be involved in any ground water deposition of the calcites, whereas it is more likely that (older) water from the deeper Paleozoic limestones and the underlying Precambrian rocks would be involved in a spring deposit produced by convecting water. While the wells at Yucca Mountain do not extend deep enough to provide samples of the water from the Precambrian, the one well that did penetrate the Paleozoic rocks gave the highest Strontium ratio measured for water at the site (.7118). The water in the Precambrian rocks, which would have been in contact with the rock for a long time, would be expected to have a very high $^{87}\text{Sr}/^{86}\text{Sr}$ ratio reflecting the high values characteristic of all Precambrian rocks. Mixing of this water

with the shallower water during convection could easily produce the values, near .7125, observed in the Trench 14 veins. Therefore, the water at Yucca Mountain would have a clear genetic relationship with hydrogenic deposits produced by convection (or seismic pumping); contrary to the conclusion by Stuckless et al. who do not seem to recognize the possibility (indeed the likelihood) of convection in an area that is geothermally and tectonically active.

Finally, Stuckless et al. use observed Uranium isotopic data to argue that since the ground water has high $^{234}\text{U}/^{238}\text{U}$ values (well above 2) and the vein calcite data they quote for Trench 14 has low values (less than 1.5), that then the calcites could not have been precipitated from the ground water at Yucca Mountain. Here they expect that, as at Devils Hole, the calcites should have Uranium ratios very close to those of the water that deposited them. However, this is only the case if the calcites exist in a closed system like that at Devils Hole. In general, as was pointed out by Hill et al. (p. 13) for example, the ^{234}U is preferentially leached from the calcite in an open system. Consequently the $^{234}\text{U}/^{238}\text{U}$ value observed under open system conditions will always be less than the value it acquired from the water at the time of deposition, and in general the longer it is exposed to open system conditions the lower the value becomes. It therefore follows that observed Uranium isotopic ratios in calcites are minimum values, the true values at deposition are always equal to, or larger than, the measured value but we can't tell for sure how much larger the depositional value might be. Stuckless et al. do not account for this possibility and so they draw an incorrect conclusion.

The proper approach is to sample the calcites extensively, seeking unleached samples, and to use the largest values measured to estimate the Uranium ratio at the time of deposition. (It will not be lower than the largest value measured.) As described by Hill et al. (see Figure 17) measured values for the calcite obtained in this way at Trench 14 indicate a value of $^{234}\text{U}/^{238}\text{U}$ at the time of deposition that was not lower than 3. This value is of the order observed for the ground water at Yucca Mountain and at Devils Hole. These new results directly contradict the conclusion drawn by Stuckless et al. and show that the Uranium isotopic data is actually compatible with a ground water source for the Trench 14 calcites.

Consequently, it is quite evident that Stuckless et al. have been led to a series of conclusions that are not justified and, in important cases, can be demonstrated to be

incorrect. Nevertheless, as is the case with Quade and Cerling's work, their conclusions have been adopted by many as accepted facts, producing perceptions that a body of evidence exists that supports a pedogenic origin for the Yucca Mountain vein calcites while, in fact, it does not.

A recent important example of misrepresentation is contained in the paper by Roedder *et. al.*, 1994, in which the authors have ignored data and results from their own previous work. In particular, these authors state: "The presence of all-fluid inclusions, and the absence of two-phase, liquid + vapor, inclusions in the upper thousand feet of the USW G-1 borehole together establish that these calcites have formed at low temperatures, <100°C, possibly comparable to modern ambient temperatures." However, we have received, through a request by the State of Nevada to the DOE, a copy of the fluid inclusion data and results that are the basis of the Roedder *et. al.* paper (but are not tabulated in the paper) and find that these authors actually determined a homogenization temperature from a calcite sample in USW G-1 at a depth of about 200 meters (669.2 ft). The only way that this determination can be made is with a two-phase inclusion. Therefore the statement that such inclusions do not occur above the 1000 ft level is false, based on their own data. Furthermore, the temperature measured by them was 81°C. Since the known geothermal gradient at Yucca Mountain is about 22°C per km (Sass, 1980), then the modern ambient temperature at the sample depth would be about 20°C. Unless they think that a factor of four discrepancy is ignorable, then the second part of their statement is also clearly false; that is the calcites in question could not, even possibly, have formed at temperatures comparable to modern ambient temperatures. Indeed this deposition temperature, at this depth, is what would be expected for an epithermal deposit associated with upward ground water movement in a faulted and highly fractured rock. In addition, they also do not mention other homogenization temperature data that they obtained in USW G-2 at shallow depths near 1000 ft below the surface and well above the present water table; in particular the data for two-phase inclusions in calcite giving temperatures of about 58°C at 858 ft (262 m), 81°C and 72°C at 1138 ft (347 m) and 104°C at 1170 ft (357 m). All these temperatures are well above modern ambient temperatures at these depths (by factors of from about 3 to well over 4) and again strongly support an epithermal interpretation. Given the Uranium series age dates for calcites from veins in these wells, many of which are as young as 30,000 to 100,000 years bp, then it would be more reasonable to infer that recent epithermal activity has occurred and could reoccur to cause repository flooding, than to conclude that these deposits are pedogenic as is implied by Roedder

et. al. This is particularly the case if fluid inclusion data from other sources (our own included) are taken into account, since many of these yield moderate to high depositional temperatures at shallow depths (and at the present surface) in calcites that have very young ages.

Because the determinations of fluid inclusion temperatures in calcite veins at the surface and at depth are so important in the interpretation of the tectonic and geologic history of the site area, we asked an outside expert to review the Roedder et. al. paper and the relevant data. Dr. Yuri V. Dublyansky, a noted Russian expert, agreed to do such a review and to provide a report. We have enclosed his report, along with copies of the Roedder et. al. paper and the pertinent data transmitted to us by the DOE, since we think your committee may be interested in his independent, more detailed, assessment as well as in the comments we have made. In any case, given the contents of the report by Dublyansky and our own analysis, it seems to us that these authors, whether by chance or design, have ended up misrepresenting their own data and results which then leads to an erroneous conclusion (namely that the vein calcites are of pedogenic origin).

A clear and important example of misrepresentation of published results that has led to erroneous perceptions (not to mention erroneous conclusions), occurs in the National Academy NRC Panel report: "Ground Water at Yucca Mountain: How High Can It Rise?" Rather than discuss the details of this issue in the body of this letter, which has grown too long as it is, a review containing a discussion of the issue, and several others, is enclosed. (Review of the NAS/NRC Report: "Groundwater at Yucca Mountain: How High Can It Rise?", by C. B. Archambeau, 1992.) This report was sent to Dr. Frank Press, President of the National Academy, as well as to the State of Nevada as a special contract report. An extensive correspondence with Dr. Press and his staff ensued and copies of all this correspondence are contained in another report (Dialogs on the Yucca Mountain Controversy, by C. B. Archambeau, 1993, Special Report No. 4 to the State of Nevada) which can be made available to you if you want to pursue this further.

In summary, the perception that the calcite veins and calcretes at Yucca Mountain are of pedogenic origin is not well founded and certainly it seems evident that the issue of an upwelling ground water hazard at Yucca Mountain is not closed. Further, it appears to us that a very strong case can now be made that ground water upwelling has

occurred episodically throughout the last 10 million years and can be expected to do the same in the future. We therefore feel that it would be appropriate for your committee to review this issue in the light of all the currently available evidence.

Sincerely Yours,



Charles B. Archambeau
Chairman, TRAC-NA
Boulder, Colorado

cc: W. Ott - NRC
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PAST RESEARCH AND RESULTS AND PLANS FOR F. Y. '95

CONTRACT No. 94/96.0003

Presented to the
Nuclear Waste Project Office
State of Nevada

July, 1994

By:

**C. B. Archambeau
J. S. Szyamanski**

PART I - FRAMEWORK FOR THE RESEARCH.

- Focus and main objective of the TRAC -N.A. research.
- Understanding of the flow system.
- Testing of the sink↔source transformation hypothesis.

FOCUS AND MAIN OBJECTIVE OF THE TRAC-NA RESEARCH.

- Focus is on long-term behavior of the flow system based on : a) study of epigenetic alteration minerals, b) study of epigenetic/epithermal vein minerals, and c) numerical simulations.
- Purpose is to determine, unequivocally, whether or not the flow system: a) underwent intermittent sink \leftrightarrow source transformations with associated large-scale fluctuations of the water table; and b) whether it may undergo such transformations in the future.

CONCEPTUAL UNDERSTANDING OF THE FLOW SYSTEM.

- A variety of geotechnical data was interpreted by the USGS to indicate that: a) the area is underlain by "a complex hydrothermal circulation system", and b) "the height of water table may be controlled by the state of stress".
- Accepting both of these conditions as being correctly inferred, TRAC-NA further concludes that: a) the conductivity structure is controlled by the state of in-situ stress and, therefore, is space-and time-dependent, and b) space-and time-depend changes of fluid and heat fluxes are intrinsic features of the system.
- Consequently, the flow system must be regarded as a non-linear system for which $\text{sink} \leftrightarrow \text{source}$ transformations constitute an intrinsic behavioral characteristic.
For such a system, the usual linear, noninteractive hydrodynamic methods that are typically employed to infer patterns of long-term behavior, are not applicable, except in very limited circumstances (e.g. equilibrium limits).

TESTING OF THE SINK↔SOURCE TRANSFORMATION

HYPOTHESIS.

- A sink↔source transformation must be accompanied by a large influx of hypogene fluids into the vadose zone and partial inundation of this zone.
- The hypogene fluids are expected to be : a) hot or warm, depending upon rate of influx, b) Ca + Mg in bulk composition, c) enriched in trace elements, and d) carrying isotopic signatures indicative of prolonged exchange reactions with the basement rocks.
- Fossil sink↔source transformations are (therefore) expected to be evidenced by: a) calcium-magnesium metasomatism (relative to the parent glass), b) fracture -based calcic zeolitization, and c) intermittent emplacement of epigenetic veins whose thermal and isotopic characteristics are consistent with those expected for the parent hypogene fluids.

PART II - TOPICS OF PAST RESEARCH.

- Chemical composition of the vadose zone tufts, relative to the parent glass.
- Chemical, mineralogic, and radiometric characteristics of epigenetic alteration minerals, from above and below the water table.
- Origin of epigenetic vein minerals, from the vadose zone, based on : a) INAA abundance patterns, b) homogenization temperatures of fluid inclusions, and c) isotopic characters of incorporated uranium, lead, strontium, oxygen, and carbon.
- Quantitative numerical simulations and formulations of hydro-tectonic phenomena: seismic pumping and convection due to earthquakes, hydrofracturing in tectonically stressed pre-fractured Medina, steady state approximations for the current water table and its high gradient near Y.M. , effects of earthquakes on the planned exploratory tunnel.

RESULTS - MAJOR ELEMENT COMPOSITION OF THE TUFFS.

TRAC-NA's analyses of chemical composition of the tuffs, from the vadose zone, yielded three principle conclusions:

- Relative to the stratigraphically equivalent glasses, the altered tuffs are: a) 0.5 ~ 5.0% enriched in alumina, b) 0.25 - 1.75% enriched in magnesia, c) 0.5 - 5.0% enriched in lime, d) 0.5 - 4.5% enriched in iron oxide, e) 0.1 - 0.5% enriched in titania, f) 4 - 10% depleted in silica, g) 0.5 - 2.5% depleted in soda, and h) 0.5 - 3.0% depleted in potash.
- The evident open system chemical alteration, or metasomatism, must be attributed to fluids that have acquired their dissolved content from outside the tuffs of Yucca Mountain.
- Known chemical composition of fluids infiltrating through the vadose zone together with the spatial heterogeneity of metasomatism indicate that, it is unreasonable to attribute the metasomatism to infiltrating meteoric precipitation. In contrast, chemical metasomatism is a feature that commonly is regarded as a product of hydrothermal metamorphism.

RESULTS - EPIGENETIC ALTERATION MINERALS.

TRAC-NA's analyses of epigenetic alteration minerals, occurring both above and below the water table, yielded three principal conclusions:

- At Yucca Mountain, two chemically and radiometrically distinct assemblages of the alteration minerals are present.
- The older assemblage is represented by alteration minerals produced through interaction of Na-K (alkalic) fluids with vitric tuffs. This epigenetic assemblage consists of : a) higher-grade montmorillonite clays (allevardite, kalkberg, and illite), b) predominantly sodic-potassic clinoptilolites, and c) higher-grade minerals from the alkali zeolitic series, specifically analcime and albite. The corresponding minerals, both montmorillonites and zeolites, carry the same K/Ar ages, ranging from 11 to 9.5 Ma B.P.. Clearly, the older assemblage represents hydrothermal metamorphism supported by the Timber Mtn. heat source.

- A later (overprinting) assemblage is represented by alteration minerals produced through interaction of Ca-Mg (alkaline earth) fluids with vitric tuffs. This epigenetic assemblage consists of: a) lower grade minerals from the calcic zeolitic series (clinoptilolite-heulandite, erionite, stellerite, chabazite), and b) higher grade minerals from the calcic zeolitic series, specifically laumontite and analcime. The corresponding alteration minerals (clinoptilolite-heulandite) carry K/Ar ages ranging from 8.5 to as little as 2.0 Ma B.P.. Chemically, the overprinting assemblage is equivalent to the whole-rock metasomatism. Both of these alteration elements record the same alteration processes.

RESULTS - INAA ELEMENTAL (36) ABUNDANCE PATTERNS.

TRAC-NA's analyses of the INAA patterns, obtained from samples of the surficial calcite-silica-sepiolite deposits, yielded four principal conclusions:

- Relative to A-horizon (vesicular) soils, the surficial deposits are significantly enriched in Co, Ni, Br, As, Sb, W, Au, and U.
- Relative to known surficial deposits of groundwater origin, the deposits yield similar elemental concentrations and abundance patterns.
- Relative to veins from below the water table, the deposits yield similar REE abundance pattern.
- Because of these observations and in contrast to the DOE's conclusions, the INAA results do not favor the pedogenic - *per descensum* origin of the calcite-silica -sepiolite deposits. Instead, these results may be regarded as supporting the competing pedogenic - *per ascensum* origin.

RESULTS - FLUID INCLUSION STUDIES.

TRAC-NA's analyses of fluid inclusion data, obtained from samples of the calcite -silica veins from the vadose zone, yielded three main conclusions:

- Contradicting DOE's statements, the calcite-silica veins (pull apart fault and boreholes USW G-1, G-2, and GU-3) contain two -phase (vapor and liquid) inclusions. Upon heating, the inclusions yielded T_{homo} ranging from 57 to as much as 227°C. Importantly, the same samples carry isotopic signatures (carbon, strontium, uranium, and oxygen) which are equivalent to those of the surficial calcite-silica deposits.
- Geothermal measurements, performed in the vadose zone, revealed that the in-situ ambient temperature increases from 15-20°C, at and near the topographic surface, to 30-40°C, at and near the water table.
- The large discrepancy between the depositional and ambient (contemporary) temperatures clearly indicates that the veins are products of epithermal fluids. This conclusion is secure beyond a reasonable doubt and effectively invalidates the *per descensum* interpretations of the origin of most of the disputed calcite-silica deposits and veins.

SUMMARY RESULTS - ISOTOPES OF URANIUM, LEAD, STRONTIUM, OXYGEN, AND CARBON.

TRAC-NA's analyses of all of the available isotopic data, derived from samples of the calcite-silica deposits/veins from the vadose zone, yielded two summary conclusions:

- In contrast to the DOE's conclusion, the isotopic data do not require rejection of the postulated *per ascensum* origin of the deposits and veins.
- Rather than requiring and/or favoring the *per descensum* origin, the isotopic data may reasonably be interpreted as either : a) requiring the *per ascensum* origin based on the results from the observed isotopes of oxygen and uranium; or if the U and O isotopic data are discounted, b) are not uniquely indicative of either of the competing depositional origins based on the results from observed isotopes of strontium, lead, and carbon.

RESULTS - ISOTOPES OF URANIUM.

TRAC-NA's analyses of the U-series isotopic data, derived from samples of the calcite-silica deposits/veins from the vadose zone, yielded three main conclusions:

- Isotopic character of the incorporated uranium is described by values of $^{234}\text{U} / ^{238}\text{U}$ ratio ranging from the equilibrium value of 1.0 to about 2.0. If it is assumed that the samples behaved as a closed system, then it can be concluded that these samples have precipitated either: a) from young meteoric fluids, (residence in a bedrock was not sufficiently long to acquire uranium isotopic disequilibrium via the α -recoil mechanism), or b) from strongly thermal fluids (where thermally stimulated fluid \leftrightarrow bedrock ionic exchange reactions annealed the α -recoil induced isotopic disequilibrium).

- Most of the analyzed samples, however, were known to have behaved as open systems. In this case values of $234\text{U} / 238\text{U}$ ratio are minimum values which were acquired via combined action of : a) the usual radioactive $234\text{U} \rightarrow 230\text{Th}$ decay, and b) preferential open system leaching of 234U atoms relative to 238U atoms. This dual control of the $234\text{U} / 238\text{U}$ ratio leads to values for the ratio that are lower than those that would occur in a closed system, so that a low uranium ratio value (at any calcite age) is not necessarily indicative of that of the parent fluid.
- A few carefully selected samples (to eliminate the undesired open system behavior) from the Trench 14 vein, however, yielded values of $234\text{U} / 238\text{U}$ ratio that are identical to those from the Devil's Hole vein. This observation, together with the fact that isotopic character of strontium incorporated in both of the veins is also identical, leads to a certain conclusion: at both locations, depositional processes and parent fluids were similar. (In the case of the DH-2 vein, the *per ascensum* origin is known with certainty so the inference would be that the TR-14 vein origins are *per ascensum* as well.)

RESULTS - ISOTOPES OF OXYGEN.

TRAC-NA's analyses of the isotopic character of oxygen, incorporated in samples of the controversial calcite-silica deposits/veins, yielded three main conclusions:

- For co-existing opal - calcite samples, the $\Delta^{18}\text{O}_{\text{opal-carb}}$ values range from 4.8 to 9.7 per mill smow (samples from the topographic surface) and from 2.30 to 6.70 per mill smow (samples from a depth ranging from 85.2 to 236.7 m). The corresponding depositional temperatures range from 20 to 60°C, for the surficial samples, and from 25 to 225°C, for the subsurface samples. These depositional temperatures are in agreement with the 57-227°C range inferred based on homogenization temperatures of the two - phase fluid inclusions. There is no reason, therefore, to regard either of the data sets as erroneous.

- The rate of decrease of the $\delta^{18}\text{O}$ values as a function of depth, $d\delta^{18}\text{O}/dz$ gradient, ranges from 7.0 to as much as 25 per mill snow/per 1 km. For samples representing the same paragenetic assemblage, the depthward decreasing values of $\delta^{18}\text{O}$ ratio are commonly regarded as reflecting paleo-geothermal gradients. For Yucca Mountain, the reconstructed value of these gradients ranges from 35 to as much as $140^\circ\text{C}/\text{km}$.
- Relative to the spatially corresponding contemporary geothermal conditions, both the depositional temperatures and the paleo-geothermal gradients are substantially higher. The discrepancies confirm the epithermal origin of the calcite-silica veins, as deduced independently based on the INAA, fluid inclusion, and U-series data.

RESULTS - ISOTOPES OF STRONTIUM.

TRAC-NA's analyses of Sr isotopic data, derived from samples of the calcite-silica deposits/veins from the vadose zone, yielded three conclusions:

- Relative to both eolian dust (vesicular A horizon) (0.71166) and pedogenic coatings (0.71145), the calcite-silica deposits/veins are noticeably enriched in radiogenic ^{87}Sr (0.7124). This isotopic discord, together with the other data (i.e. INAA, fluid inclusion, U-series, and isotopes of oxygen), casts doubt on the DOE's interpretations of the *per descensum* origin of these deposits/veins.
- In contrast, the calcite-silica deposits/veins carry values of $^{87}\text{Sr} / {^{86}\text{Sr}}$ ratio that are similar to those from: a) metasomatically altered host bedrock, and b) epithermal veins from the Devil's Hole cavern. This strontium isotopic affinity favors the competing *per ascensum* origin and further reinforces the independently inferred epithermal origin of these deposits/veins.

- The radiogenic ^{87}Sr that is incorporated in both the metasomatically altered and hydrothermally mineralized bedrock (tuffs and the underlying Paleozoic carbonates) and the calcite-silica veins from the vadose zone, was undoubtedly derived from clastic metasediments comprising the Precambrian basement. What is debatable based on the strontium data alone, however, is a mode of transport of the radiogenic strontium from the Precambrian basement. Because both the Holocene accumulations of eolian dust and the contemporary groundwaters do not carry the sufficiently radiogenic strontium, the strontium isotopic data alone are of limited assistance in that regard.

RESULTS - ISOTOPES OF LEAD.

TRAC-NA's analyses of Pb isotopic data, derived from samples of both the host bedrock and the calcite-silica deposits/veins, yielded two conclusions:

- Lead, that is incorporated in both the metasomatically altered tuffs and the calcite -silica deposits/veins from the vadose zone, has been derived from metasediments comprising the underlying basement. In both of these cases, the lead isotopic ratios ($^{206}\text{Pb} / 204\text{Pb}$) are adequately accounted for by postulating that the lead was derived through mixing of leads incorporated in both of the fractions (leachate and residuum) comprising the basement rocks.
- Similarly, as is the case with the strontium isotopic data, the lead isotopic data do not provide a clue on how the lead was transported from the basement into the vadose zone. Although not contradicted by the lead isotopic data, the USGS interpretations of the *per descensum* origin of the calcite-silica veins are not uniquely constrained and, therefore, not necessarily correct.

RESULTS - ISOTOPES OF CARBON.

TRAC-NA's analyses of isotopic character of carbon, incorporated in the calcite-silica deposits/veins from the vadose zone, yielded two main conclusions:

- Values of del¹³C ratio range from -9.0 to as much as 4.0 per mill PDB. As far as origin of the incorporated carbon in concerned, the carbon isotopic data allow for consideration of three source possibilities. These are: a) biosphere, b) magmatic source, and c) mixed source (carbon acquired through dissolution of the Paleozoic limestones is intermixed with "light" carbon of either biogenic origin or magmatic origin).
- Although not contradicted by the carbon isotopic data, the *per descensum* interpretations of origin of the calcite-silica veins/deposits from the vadose zone are not uniquely constrained and, therefore, not necessarily correct. Similarly as is the case with the strontium and lead isotopic data, the competing *per ascensum* interpretations are equally valid.

OVERALL CONCLUSION - LONG-TERM BEHAVIOR OF THE YUCCA MOUNTAIN FLOW SYSTEM.

- A large set of chemical, mineralogic, fluid inclusion, geochronologic, and isotopic data has been assembled to form a coherent picture of invasions of the vadose zone by epithermal fluids. Intermittently, these invasions occurred during the Plio-Quaternary time span and are recorded through a paragenetic assemblage consisting of three chemically and isotopically compatible (involving isotopes of strontium and lead) elements. These are: a) metasomatic alteration, b) calcic zeolitization, and c) carbonatization.
- With regard to the long-term performance of a high-level nuclear waste repository, this conclusion points-out a hazard that can not be mitigated and reduced to prudent and acceptable levels. Because TRAC -NA regards this conclusion as a pivotal point in resolving the suitability questions, most of our efforts are directed toward: a) reinforcing it, and b) informing the scientific community.

RESULTS - MODELING OF HYDROTECTONIC AND TECTONIC PROCESSES AT YUCCA MOUNTAIN. (1.)

TRAC-N.A. is developing quantitative analysis methods based on new formulations of interactive hydrodynamic-tectonic (HydroTECTonic) systems and applying them to: a) analysis of hydrological data from Yucca Mt., and b) prediction of possible episodic behavior of the water system in response to tectonic effects. In addition, tectonic effects on the exploratory tunnel, due to design level earthquakes, are also considered. Results include:

- Formulation of fluid and gas flows through fractures in a stressed solid, wherein the fractures are irregular and have boundaries that move in response to tectonic stress changes as well as fluid /gas pressure changes.
- Formulation of an analytical framework for the analysis of slug/hydrofracture data resulting from tests in a pre-fractured, tectonically stressed medium.

RESULTS - MODELING OF HYDROTECTONIC AND TECTONIC PROCESSES AT YUCCA MOUNTAIN. (2)

- Investigations of steady state flow system patterns produced by different models of the hydraulic conductivity at and near Yucca Mountain show that the water table and its steep gradient, occurring at the north end of Yucca Mountain and elsewhere in a very narrow 100 km long east-west trending zone, can be explained by variable fracture-controlled hydraulic conductivity that is in agreement with measured conductivities at wells in the region. It is also demonstrated that a moderate sized shallow earthquake in the north central area of Yucca Mt. is likely to cause a southward shift of the steep gradient and result in a large rise (150 to 300 m) in the water table due to fracture closure.
- Analysis of slug test data shows that some fractures at Yucca Mt. are open and that others open at low pressures, and in some cases at very low pressures, so that rapid flow out of the well often occurs through the open fracture conduits. The low pressure required for fracture opening and the inferred presence of pre-existing open fractures requires high tectonic stress levels and dilatant conditions.
- Preliminary modeling of the long-term deformational response of the medium, around the planned exploratory tunnel, to a design level earthquake on one of the nearby major fault zones indicates stress changes that are likely to be sufficient to produce failure at stress concentration points at and near the tunnel boundary.

**OVERVIEW OF CALCITE/OPAL DEPOSITS AT OR NEAR THE PROPOSED
HIGH-LEVEL NUCLEAR WASTE SITE, YUCCA MOUNTAIN, NEVADA:
PEDOGENIC, HYPOGENE, OR BOTH?**

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Calcite/opal deposits at Yucca Mountain were studied with respect to their regional and field geology, petrology and petrography, chemistry and isotopic geochemistry, and fluid inclusions. They were also compared with true pedogenic deposits. Some of the data is equivocal: it can support either a hypogene or pedogenic origin for these controversial deposits. However, Sr-isotope and fluid inclusion data favor a hypogene interpretation while petrographic textures favor a pedogenic interpretation. A possible resolution of this dilemma is that a pedogenic component may overprint a hypogene component. This subject relates to the suitability of Yucca Mountain as a high-level, nuclear waste site.

Introduction

This multidisciplinary study of the controversial calcite/opal deposits (CCOD) at Yucca Mountain, Nevada considers three main models of origin: the epithermal/hypogene, the groundwater/spring, and the pedogenic/supergene. All three models involve meteoric water but each invoke a different chemical and isotopic evolution for this water and precipitation of the CCOD. This study is intended to re-examine the CCOD based on additional field work and a more complete set of geochemical/geological data. It is hoped that this study will point out areas of possible future research.

The origin of the CCOD has been a matter of considerable debate because it relates to the suitability of Yucca Mountain as a high-level radioactive waste site. If the CCOD have an epithermal/hypogene origin, then upwelling water could breach the waste site in the future. It has often been stated that the geologic evidence "precludes" the involvement of upwelling waters in the deposition of the CCOD (e.g., 1, 2), but does it? As reported in *Science*, April 18, 1992, p. 247 a 17-member panel convened by the National Academy of Sciences concluded that "there is no compelling evidence for the repetitive flooding of the environment by expulsion of groundwater" and "instead, the evidence strongly supports

the idea that the near-surface mineral deposits resulted from percolating rainwater, which carried soil minerals down into rock fractures" (3). Such conclusions have led the Department of Energy to state that it "finds no basis to continue to study the origin of these specific deposits" (4). Other studies, however, have come to the opposite conclusion: that a hypogene, upwelling-flow model can explain the CCOD at Yucca Mountain (5, 6).

Regional Geology

Yucca Mountain is located in the southern Great Basin, a tectonically-active area characterized by north-trending linear mountain ranges flanked by extensive alluvial fans and separated by broad alluvial basins (Fig. 1). Quaternary volcanism, active faulting and seismicity, high heat flow, and thermal springs characterize the region. The most recent volcanic features are a series of volcanic cones in Crater Flat: Red Cone (1.0-1.5 ma), Little Cone (1.11 ma), Black Cone (1.07-1.09 ma), and Lathrop Wells Cone (119±11 to 141±10 ka) (7, 8). Faulting in the area (within 40 km of Yucca Mountain) dates from the Holocene (200-2,000 ybp) to Pleistocene (9, 10, 11). These faults trend predominantly north-south (Fig. 1), but a recently-mapped shear zone, called the Sundance fault, trends northwest-southeast through the planned waste site (12). Earthquakes are common in the area, the most recent occurring on June 29, 1992 and measuring 5.6 on the Richter scale. This earthquake, which had its epicenter near Little Skull Mountain located just 17 km southeast of the proposed high-level waste repository, caused considerable damage to the Yucca Mountain Project Operations Center on the Nevada Test Site. Heat flow is as high as 130 mWm⁻² which is significantly above Basin and Range heat-flow averages of 80 to 100 mWm⁻² (13). Thermal-spring water temperatures measure 34°C at Devils Hole and 43°C at Oasis Hot Springs (14).

The CCOD are localized along Quaternary faults recognizable in the field by offset beds, well-exposed and slickensided surfaces, or brecciated and mineralized zones. In places where faults are well-exposed, the CCOD occur primarily as sub-vertical seams or veins along or near the fault plane, and either die out away from the fault (e.g., Wailing Wall; Fig. 2) or form downslope from the faults and veins (e.g., Busted Butte; Fig. 3). A number of the CCOD are located along major faults: Trench 14 along the Bow Ridge fault; Busted Butte along the Paintbrush Canyon fault; Trench 8, New Trench, and WT-7 along the Solitario Canyon fault; Wailing Wall along the Stagecoach Road fault, and Crater Flat along the Windy Wash fault (Fig. 1).

The fault calcite/opal often displays a vertical vein morphology where exposed by trenching or valley downcutting. This vein geometry is well illustrated at Trench 14 and elsewhere, but is most dramatic on the west and east sides of Busted Butte where valley erosion has dissected sand ramps (Fig. 4 and/or Cover photo). These veins narrow towards the base but thicken and splay out into multiple veins near (within a few meters of) the sand-ramp ground surface. The CCOD then continues downslope from these feeder veins, sometimes reaching or surpassing the toe of slope of the sand ramp (Fig. 3). Such a splayed geometry is typical of epithermal mineral deposits (15).

Yucca Mountain is located between two mining districts of epithermal mineralization: Bare Mountain ~11 km to the west and Wahmonie ~20 km to the east (Fig. 1). The Wahmonie Mining District was mined primarily for silver, but also has a high concentration of cobalt, chromium, and gold (16); the Bare Mountain Mining District was mined primarily for gold, fluorite, and mercury, but also has a high concentration of arsenic, cadmium, lead, and zinc (17). This mineralization is evidence that deep hydrothermal circulation has occurred within the upper crust.

Comparison of Calcite/Opal and True Pedogenic Deposits

Four types of calcite and/or silica deposits exist at or in the vicinity of Yucca Mountain: (i) true pedogenic deposits (TPD), (ii) groundwater spring deposits (GSD), (iii) controversial calcite/opal deposits (CCOD) along faults and downslope from faults, and (iv) calcite vein deposits (CVD) in the subsurface. The main controversy revolves around whether the CCOD and CVD are of pedogenic/supergene origin as are the TPD, or if they are GSD of hypogene (deep-seated) origin and therefore a potential threat to the integrity of the proposed nuclear waste repository. Past studies supporting a pedogenic hypothesis have assumed that CCOD slope calcretes are "pedogenic" in origin (e.g., 18, 19) and, since the stable isotopic composition ($\delta^{13}\text{C}$, $\delta^{18}\text{O}$, $^{87}\text{Sr}/^{86}\text{Sr}$) of the CCOD veins matches that of the CCOD slope calcretes (Figs. 13, 15), it has therefore been concluded that these veins are "pedogenic." Furthermore, since the CVD also have similar isotopic compositions (Figs. 14, 16), they too have been assumed to be derived from infiltrating meteoric water (20).

In order to distinguish between what is truly pedogenic and what may be hypogene, it is first necessary to (i) define what is meant by "pedogenic", and (ii) establish criteria for the recognition of TPD and then to compare these with the CCOD. We define "pedogenic" to mean related to, or involved in, the development of soils, *regardless* of the source of the material being subjected to soil-forming processes. (The CCOD could be originally supergene or hypogene and then be subjected to pedogenic/supergene processes; e.g., seep areas can be colonized by vegetation and undergo pedogenesis). As established by soil scientists (e.g., 21, 22, 23), TPD display the following features: (i) they occur as calcic or petrocalcic horizons just beneath the land surface and are oriented approximately parallel to this surface, unless exhumed (Fig. 5); (ii) they are laterally continuous with geomorphic surfaces which, in many instances, cover tens to hundreds of km^2 ; (iii) they typically consist of a detrital fabric impregnated by calcite crystals; (iv) they

become progressively more complex within progressively-older geomorphic surfaces, following a six-stage sequence (stages I to VI); and (v) they accumulate slowly, with stage I carbonates in soils of late Pleistocene age (or younger) and with stage V-VI carbonates being hundreds of thousands (or millions) of years old (23).

The TPD at Yucca Mountain meet these five criteria (Fig. 5) but differ petrographically and petrologically from the CCOD in the following respects (Table 1). The CCOD consist of very fine-grained (usually $<5\text{ }\mu\text{m}$; Fig. 6A) calcite and opal ("CT" and "A"), with minor amounts of sepiolite and quartz, and trace amounts of pyrite/chalcopyrite (24). Almost invariably these are an intimate mixture of calcite and opal, with carbonate ranging from ~20-75% and silica from ~25-80%, and with calcite/opal lamination/bands on the order of mm- to cm-thick. TPD, however, contain detritus within a carbonate matrix (Fig. 6B), where opal occurs as stringers (50-100 μm) or as void, ooid, and root-structure fillings. Crystal size of this calcite ranges from $\sim 1\mu$ to as much as millimeters. The CCOD also display a variety of petrologic textures, ranging from pure (Fig. 7), mixed (Figs. 7, 8, 10), banded/laminated (Fig. 8), massive (Figs. 7, 8), powdery (Figs. 8, 9, 10), patchy, brecciated, flow (Fig. 9), vesicular/ phenocrystic (Figs. 7, 9), veined, invasive (Fig. 10), botryoidal, ooidal, root-cast (Fig. 11), and speleothemic (25). On the other hand, TPD display powdery texture, ooidal texture, small rhizoliths, but no large root casts (Table 1).

There are a number of good reasons why the vein and slope CCOD have been taken to be of pedogenic origin. Micritic texture is not usual for hydrothermal or vein calcites but is characteristic of many pedogenic carbonate deposits. Aragonite, not calcite, is the mineral that usually precipitates at hot springs (26). Sepiolite is a magnesium silicate mineral which often forms in pedogenic caliches of arid regions. Root cast texture, rhizoliths, calcified microorganisms, argillans, ooidal texture, and detrital grains are other features of the CCOD which have been considered to be pedogenic.

All of these features seem to implicate a pedogenic origin for the CCOD; however, other explanations are possible. The fact that the CCOD are fine-grained might indicate

extremely-quick cooling and/or degassing of hypogene solutions (27). The calcite/aragonite problem is a complicated subject and in near-surface environments CaCO_3 is not necessarily deposited in equilibrium, especially if there is much dissolved CO_2 (27). Sepiolite can be pedogenic but in the Basin and Range Province it is often related to hydrothermal water upwelling along fault zones where the source of Mg and Si for the mineral is dolomite and volcanic rocks in the subsurface (24, 28). Invasive texture (Fig. 10) and patchy texture suggest penetration of later fluids of slightly different composition after sections of the calcite/opal matrix had either solidified or partly solidified. Vesicular texture (Fig. 9) is suggestive of gas cavities created by the degassing of fluids out of which the CCOD precipitated. Root-cast texture (Fig. 11) may indicate hypogene waters exiting from springs which may have supported vegetation (trees, plants), and detrital grains might have accumulated at spring orifices by eolian and/or gravitational processes, especially in sand ramps. A modern hydrothermal spring which displays root-cast texture, sand, and fossilized bacteria is Tecopa Spring, located ~90 km south of Yucca Mountain (29).

Chemistry

A question of major importance is: What is the source of calcium for the calcite fraction of the CCOD? The volcanic-tuff host rock is calcium-poor (<3 wt%), so it has been argued that the calcium for the CCOD was supplied by eolian calcareous dust source (the pedogenic or "per descensum" model) (2) or, alternatively, by Paleozoic carbonate-Precambrian rock (the hypogene or "per ascensum" model) (6). Critical to this question is whether or not there is volumetrically enough calcareous dust at Yucca Mountain to have supplied the large amount of calcium contained in the CCOD (24).

Trace element correlations (31 elements, including REE's) from a total of 143 samples for the CCOD, GSD, TPD, and soils at or near Yucca Mountain have produced equivocal

results (4, 30). Both the vein and slope CCOD at Trench 14 were found to be similar in their elemental profile and in their enrichments in Co, Ni, Br, As, Sb, W, Au, and U relative to detrital soil; i.e., the vein and slope calcrites have both derived from the same source. (The question is: are they *both* pedogenic or are they *both* hypogene?). A Fe (%)/Sc (ppm) ratio of 0.322 ± 0.016 in five Trench 14 A-horizon soils agrees with (4) and would seem to favor a pedogenic origin for these deposits. However, a Fe/Sc ratio of 0.28 ± 0.08 for 12 Cambrian Bonanza King carbonates overlaps with the vein- and slope-calcrite ratios and *alternately* can be considered to favor a hypogene mechanism (30).

Similar elemental enrichment patterns for the CCOD, GSD, and TPD also suggest that similar chemical and physical processes were operative in the genesis of all three types of deposits (Fig. 12). However, significantly higher enrichments of Co, As, Sb, W, Au, and U in the CCOD and GSD over the TPD favors a spring origin over a pedogenic one. Relatively-high concentrations of zinc in the CCOD at Trench 14 (210 ppm, 130 ppm) (4), Trench 8 (166 ppm), New Trench (90 ppm) and Wailing Wall (90 ppm), and epigenetic quartz containing grains and microveinlets of pyrite/chalcopyrite at WT-7, Wailing Wall, and Pull Apart fault, also favor an epithermal origin for the CCOD (24).

Isotope Geochemistry

Strontium Isotopes - Strontium isotopes are important indicators of the ultimate source or sources from which Sr in aqueous solution is derived and as such have been used extensively in the study of the CCOD (1, 2, 31, 32, 33, 34, 35). Strontium ratios ($^{87}\text{Sr}/^{86}\text{Sr}$) for all reported Yucca Mountain samples (surface and subsurface) are plotted in Fig. 13 (36). Significant trends are:

(i) The vein and slope CCOD have high $^{87}\text{Sr}/^{86}\text{Sr}$ ratios which fall within a narrow range and which have remarkably-constant averages (0.71221-0.71240). This implies that

vein and slope calcretes derived from the same source. A few samples have much lower ratios (dots; Trench 14, Busted Butte, Site 106).

(ii) GSD have a somewhat higher range of $^{87}\text{Sr}/^{86}\text{Sr}$ ratios than the CCOD (avg. = 0.71228-0.71306), although it overlaps with the CCOD. One data point each for the Diatomaceous Earth and Wahmonie Mound sites are much lower.

(iii) TPD have a somewhat lower range of $^{87}\text{Sr}/^{86}\text{Sr}$ ratios than the CCOD (0.71069-0.71231), although it overlaps with the CCOD. This range is similar to other pedogenic coatings and cements in the Yucca Mountain region (0.7108-0.7124) (37). The lower TPD values raise the question: Might the low- ^{87}Sr samples from Trench 14, Busted Butte, and Site 106 (and also the Diatomaceous Earth and Wahmonie Mound sites) represent a true pedogenic end member of a mixed pedogenic/hypogene system?

(iv) The fact that the Wailing Wall, WT-7, Pull Apart fault, and Bare Mountain calcite/opal plots within the range of the CCOD is important. Quartz and pyrite/chalcopyrite are minor constituents at the Wailing Wall, WT-7, and Pull Apart fault; high fluid inclusion temperatures ($T_h = 147^\circ\text{C}$, mean) have been measured at Pull Apart fault; and the Bare Mountain calcite/opal was collected along a fault within the Bare Mountain Mining District (24). An epithermal origin thus implied for these four deposits further implies that all of the isotopically- and texturally-similar CCOD at Yucca Mountain have an epithermal origin.

(v) Paleozoic carbonate and Tertiary volcanic rock in the Yucca Mountain region have elevated $^{87}\text{Sr}/^{86}\text{Sr}$ ratios and have been significantly altered over "normal" rock (34). This is especially true for the Black Marble Hill and Bare Mountain areas. This alteration has been attributed to hydrothermal solutions (38).

(vi) $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of CVD increase from the saturated zone (Fig. 14, A), to near the water table (B), and then to the unsaturated zone (C) and the CCOD (D). The low ratios (0.709-0.710) of Group A calcites reflect deep-seated, rock-water interactions with unaltered Paleozoic carbonates (20, 32). Group B ratios, which plot near the mean

Cenozoic ground water (vertical arrow), may indicate a time when the water table was higher (~85 m) than it is today (20, 33). The similar Sr-isotopic character of Groups C and D implies that both the calcite in the unsaturated zone (down to ~400 m) and the calcite in the vein and slope CCOD is derived from the same source.

Four possible sources of strontium exist for the CCOD at Yucca Mountain: (i) eolian calcareous dust, (ii) Tertiary volcanic rock, (iii) Paleozoic carbonate rock, and (iv) Precambrian rock. The eolian interpretation (2, 37) seems unlikely because eolian dust is not sufficiently enriched in ^{87}Sr to be the only source of strontium for the CCOD (Fig. 13). However, it could have been a partial source (one end-member) because the highest eolian values are equivalent to the lowest CCOD values. Both the Sr-isotope ratios and Ca-content of Tertiary volcanic rock are too low to have supplied the strontium and calcium for the CCOD; however, it could have supplied the silica for the opal. Paleozoic carbonate rock also cannot be the entire source of strontium to these deposits because neither the unaltered or altered limestone (except the highest values at Black Marble Hill and Bare Mountain) have appropriately high Sr-isotope ratios (19, 38).

The only source capable of supplying the high Sr-isotope ratios characteristic of the CCOD is Precambrian rock ($^{87}\text{Sr}/^{86}\text{Sr} = 0.7703-0.8878$ + Late Proterozoic rock, Bare Mountain; Fig. 13). A situation can be envisaged where Precambrian rock-water interactions at depth import a high $^{87}\text{Sr}/^{86}\text{Sr}$ character to the water; then, as convecting geothermal fluids flow through the above-lying Paleozoic carbonate and Tertiary volcanic aquifers, they pick up calcium and silica and acquire lower Sr-values; and finally, the calcite/opal precipitated along faults from this ascending water is reworked by surface processes so as to exhibit a pedogenic overprint. This model explains the large range of Sr-isotope ratios of springs and wells in the Yucca Mountain region ($^{87}\text{Sr}/^{86}\text{Sr} = 0.7082-0.7279$), which implies a more radiogenic source of Sr than the regional carbonate system can supply (19), and it is also consistent with the Sr-isotope, rock-water interaction model of (39). The Sr-isotope data, rather than "precluding" the involvement of ascending water

(2), favors a past to present involvement of the Precambrian basement in the ground water system *and* in the formation of the CCOD.

Carbon-Oxygen Isotopes

The stable isotopes of carbon and/or oxygen are useful as geochemical tracers and geothermometers, and have also been applied to the CCOD (18, 40, 41). C- and O-isotopic compositions for all reported surface and subsurface calcites are plotted in Figs. 15, 16 (36). Significant trends are:

(i) The vein and slope CCOD all plot within the same range of C- and O-isotope values (Fig. 15) suggesting that all derived from the same source. Calcite/opal at the Wailing Wall, WT-7, Pull Apart fault, Bare Mountain, and along the Eleana thrust fault (located just north of Yucca Mountain; ETR, Fig. 15) also plots with the CCOD suggesting (as did the Sr-isotopes) an epithermal origin for all of the CCOD. Data points for the Busted Butte, Site 106, and Trench 8 vein and slope calcrites seem to define sub-linear, evaporation and/or CO₂-loss trends, whereas the Trench 14 calcrites do not.

(ii) GSD either plot with the CCOD (WM, Fig. 15) or they are somewhat enriched in ¹³C (199, Fig. 16).

(iii) The stable isotope composition of TPD (*, Fig. 15) varies as a function of elevation; i.e, there is an overall decrease in $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values with increasing elevation, as is expected for pedogenic deposits (e.g., 42). Despite similar elevation differences for the CCOD no such trend exists for these deposits.

(iv) Paleozoic carbonaté rock (LS, Fig. 16) has been altered with respect to "normal" limestone ("unaltered LS/DOL," Fig. 16) as it was in its Sr-isotopic composition.

(v) C- and O- isotopes, like the Sr-isotopes, fall into four categories corresponding to the saturated zone (A), the shallow-phreatic zone near the water table (B), the unsaturated zone (C), and the surface CCOD (D), respectively (Fig. 16). Calcites within

Group A show the influence of increased temperature with depth (progressively-lower $\delta^{18}\text{O}$ values) and of increased fluid interaction with unaltered Paleozoic carbonate rock (progressively-higher $\delta^{13}\text{C}$). Calcites in Group B plot with the Devils Hole calcite as is consistent with the shallow-phreatic position of both. Calcites in the unsaturated zone (C) overlap significantly with the CCOD (D), especially the calcites from drill hole USW G-3/GU-3.

The C- and O-isotope data are equivocal: they can be interpreted as being due to soil processes related to vegetation type and paleoclimate during the last full-glacial (16-19 ka) (18), or they can be interpreted as being due to a hypogene mechanism (6). Calcites formed under non-equilibrium conditions could display the observed range of $\delta^{13}\text{C}$ of -9 to -3‰: both isotope systems can be affected by kinetic isotope fractionations if carbonate deposition occurs by a non-equilibrium process such as rapid CO_2 degassing (6). The problem with the vegetation/paleoclimate scenario (18) is that the dates on the CCOD (30 to >400 ka; Fig. 18) cover several late Quaternary glacial and interglacial periods and, therefore, such calcites should vary widely in their isotopic composition. The problem with the mainly-hypogene scenario (6) is that the $\delta^{18}\text{O}$ values are nearly identical to those predicted for carbonate precipitated from meteoric water at low temperatures.

Lead Isotopes

Lead isotope ratios ($^{206}\text{Pb}/^{204}\text{Pb}$; $^{208}\text{Pb}/^{204}\text{Pb}$) are also equivocal and, alternately, have been described as being "pedogenic...from eolian dust" (43) or "from clastic sedimentary, igneous or metamorphic crystalline rocks" (44). Paleozoic and Precambrian rocks contain lead with an isotopic composition that strongly suggests they are a major source of lead to the CCOD (44). It is possible that this lead could have been supplied indirectly through eolian dust, but since few of these rocks are exposed at Yucca Mountain, it is perhaps more likely that this lead was supplied directly by ascending water.

Uranium Isotopes

Uranium isotopes have been used: (i) to determine the activity ratios ($^{234}\text{U}/^{238}\text{U}$) of the calcite/opal (Fig. 17), and (ii) to date these deposits by the U-series disequilibrium method (Fig. 18). Activity ratios can be used to estimate parent fluids for deposits behaving as *closed systems*. For a coarse-grained deposit submerged in a fluid saturated with respect to calcite (e.g., Devils Hole, Fig. 17) the assumption of closed-system behavior seems appropriate, but in the vadose zone meteoric water could have removed ^{234}U by the "alpha recoil" mechanism and by leaching from radiation-damaged sites (5). In addition to these "uranium leaching" effects, some of the CCOD samples have been contaminated by detrital thorium. Both of these factors can lead to "open system" behavior: producing $^{230}\text{Th}/^{234}\text{U}$ ratios >1.0 -1.2, or resulting in $^{230}\text{Th}/^{234}\text{U}$ ratios <1.0 but still higher than they would be for a completely closed system. Also for an open system, measured $^{234}\text{U}/^{238}\text{U}$ ratios represent minimum value estimates (unleached values would be higher): therefore, low values do not necessarily indicate a pedogenic origin, as concluded by (2) or as shown by (45), but may also characterize spring deposits (Fig. 17, Amargosa Basin and Furnace Creek Wash). In addition, it is important to realize in the context of possible pedogenic reworking of the CCOD, that open-system behavior may result in *younger* ages for these deposits than has been determined under assumed closed-system conditions (5) (24) (46).

New results for carefully-selected CCOD samples, corrected for detrital thorium, are compared in Fig. 17 with older "closed system" data from Devil's Hole. Four samples had $^{230}\text{Th}/^{234}\text{U}$ ratios >1.2 and were unfit for chronometric dating (not shown on Fig. 17), while the other four appear to have remained relatively closed systems (*, Fig. 17). The results from Trench 14, Busted Butte, and also from southern Crater Flat (45) show $^{234}\text{U}/^{238}\text{U}$ activity ratios that are distinctly higher than those expected to be associated

with pedogenic deposits and instead are consistent with precipitation from deep ground waters.

The age data (Fig. 18) are important, even though they should be considered maximum ages for the reasons discussed above. First, they show that some of the CCOD are very young (~30 ka; Fig. 18) and are thus unlikely to be part of pedogenic horizons which can take hundreds of thousands of years to form. Second, they show that carbonate deposition has occurred over the last 400 ka and that there may have been discrete depositional episodes at c. 20, 30, 40-50, 70-80, 90, and 110 ka (and perhaps at 180 ka and 250-270 ka). Such episodes favor a discontinuous hypogene-pumping mechanism over a more-continuous pedogenic process. Episodic activity may even possibly relate to the current controversy surrounding Devils Hole and the Milankovitch theory (47, 48) and to the "paleoclimatic riddle" of (49): isotopic changes involved with this activity could be offsetting the climatic record.

Fluid Inclusions

The extremely fine-grained nature of the CCOD (Fig. 6A) precludes fluid inclusion measurements. However, fluid inclusion temperatures have been obtained on epigenetic quartz at Pull Apart fault ($T_h = 147^\circ\text{C}$, mean) (46) and on more coarsely-crystalline subsurface CVD (4, 50). These fluid inclusion temperatures are extremely important to the pedogenic-hypogene debate:

- (i) Two-phase inclusions with elevated filling temperatures *cannot* be formed by pedogenic processes.
- (ii) High-temperature calcite is found at relatively shallow depths; e.g., calcite at a 31 m depth in USW G-3/GU3 has $T_h = 101-227^\circ\text{C}$ (36). Elevated temperatures are not compatible with current geothermal gradients and show no relationship to depth (29).

(iii) C-, O-, and Sr-isotope ratios of CVD from the same depth as calcites with high fluid inclusion temperatures plot within the field of the CCOD. For example, the 31 m-deep USW G-3/GU3 calcite has a $\delta^{13}\text{C} = -6.4\text{\textperthousand}$, $\delta^{18}\text{O} = -9.4\text{\textperthousand}$ which corresponds to Trench 14 CCOD ratios (Fig. 15).

(iv) Age of calcite from the same depth as calcites with high fluid inclusion temperatures can be very young. For example, calcite at 131 m depth in USW G-3/GU-3 ($T_h = 125\text{-}170^\circ\text{C}$) has a U-series age of 26 ± 20 ka (36).

The pertinent questions to ask are: How can so-called "pedogenic" or "supergene" deposits (20) have such high fluid inclusion temperatures? And, if these deposits are very young and of hypogene origin, might not upwelling water pose a threat to the proposed waste repository site?

Discussion

Some of the petrographic features of the CCOD at Yucca Mountain support a pedogenic origin: the presence of calcite instead of aragonite; micritic, root-cast, and ooidal textures; and detrital grains. Much of the data are equivocal: sepiolite, trace element correlations, carbon-oxygen and lead isotopes. But some of the data favor a hypogene origin: petrologic features such as flow, invasive, patchy, and vesicular texture; pyrite/chalcopyrite; elemental enrichment patterns; strontium and uranium isotopes; and fluid inclusion temperatures.

How can this dilemma be resolved? A pedogenic overprint of an epithermal/hypogene component could account for the data. If deep-seated water had ascended along faults, it would have carried the chemical and isotopic signature of Precambrian, Paleozoic, and Tertiary rock through the unsaturated zone to the surface thus creating the CVD and CCOD both with the same C-, O-, and Sr- isotopic composition. The surficial CCOD would then have been reworked by pedogenic/supergene processes including plant and

microbial activity, leaching, and isotopic resetting, thus giving the deposits the *appearance* of being totally pedogenic.

The very fact that the CCOD are characterized by a broad suite of equivocal data and contradictory possible interpretations should, along with the fact that the site is in an extremely active tectonic area, be a red flag of caution that warrents the most conservative-possible stance with respect to the placement of a high-level radioactive waste site at Yucca Mountain.

REFERENCES AND NOTES

1. B. D. Marshall and S. Mahan, *Geol. Soc. Am. Abstr. Prog.*, 23, A118 (1991).
2. J. S. Stuckless, Z. E. Peterman, D. R. Muhs, *Science*, 254, 551 (1991).
3. National Academy of Sciences Panel, *Ground water at Yucca Mountain: How High Can it rise?* (Natl. Acad. Press, Washington, DC, 1992).
4. Department of Energy, *Report YMP193-II-R* (U.S. Dept. Energy Project Office, Las Vegas, NV, 1993).
5. C. B. Archambeau and M. J. Price, *Minority Report, Special DOE Review Panel* (U.S. Dept. Energy, Las Vegas, NV, 1991).
6. J. S. Szymanski, *U.S. DOE Report, April, 1992* (U.S. Dept. Energy, Las Vegas, NV, 1992).
7. S. G. Wells, L. D. McFadden, C. E. Renault, B. M. Crowe, *Geol.*, 18, 549 (1990).
8. B. D. Turrin and D. E. Champion, *Proc. High Level Radioactive Waste Management Conf.* (Am. Nuclear Soc., La Grange Park, IL, 1991), p. 68-75.
9. M. C. Reheis, *U.S. Geol. Surv., Open File Rep. 86-576* (1986).
10. G. E. Brogan, K. S. Kellogg, C. L. Terhune, *U.S. Geol. Surv. Bull.*, 1991 (1991).
11. C. M. Menges *et al.*, *Proc. Int. High Level Radioactive Waste Conf.*, May 23-27, (Am. Soc. Civil Engineers - Am. Nuclear Soc., Las Vegas, NV, 1994), p. 2373-2386.

12. R. W. Spengler, C. A. Braun, L. G. Martin, C. W. Weisenberg, *U.S. Geol. Surv., Open File Rep. 94-49* (1994).
13. J. H. Sass, A. H. Lachenbruch, E. W. Mase, *U. S. Geol. Surv., Open File Rep. 80-826* (1980).
14. I. J. Winograd and F. J. Pearson, *Water Resources Res.*, **12**, 1125 (1976).
15. B. R. Berger and P. Eimon, *Soc. Mining Eng., Reprint 82-13* (1982).
16. J. Quade and J. V. Tingley, *Rept. DOE/NV/10295-1* (Nevada Bur. Mines, Reno, NV, 1983).
17. J. V. Tingley, *Rept. 39* (Nevada Bur. Mines, Reno, NV, 1984).
18. J. Quade and T. E. Cerling, *Science*, **250**, 1549 (1990).
19. B. D. Marshall *et al.*, *Proc. Int. High Level Radioactive Waste Management Conf.*, April 8-12 (Am. Soc. Civil Engineers-Am. Nuclear Soc., Las Vegas, NV, 1990), p. 921-923.
20. Z. E. Peterman, J. S. Stuckless, B. O. Marshall, S. A. Mahan, K. Futa, *Proc. Int. High Level Radioactive Waste Management Conf.*, April 12-16 (Am. Soc. Civil Engineers-Am. Nuclear Soc., Las Vegas, NV, 1992), p. 1582-1586.
21. L. H. Gile, F. F. Peterson, R. B. Grossman, *Soil Science*, **101**, 347 (1966).
22. L. H. Gile, J. W. Hawley, R. B. Grossman, *Guidebook to the Desert Project* (New Mexico Bur. Mines Min. Resources, Socorro, NM, Mem. 39, 1981).
23. M. N. Machette, in *Soils and Quaternary Geology of the Southwestern United States*, D. L. Weide, Ed. (Geol. Soc. Am. Spec. Paper 203, 1985), p. 1-21
24. C. A. Hill *et al.*, in *Annual Report-Nevada*, D. E. Livingston, Ed. (Technology Resource Assessment Corp. Rept. to Nuclear Waste Project Office, Carson City, NV, January, 1994).
25. For a discussion and additional photos of textural types refer to (24) and also to C. A. Hill and C. M. Schluter, *Special Report no. 7* (Technology Resource Assessment Corp. Rept. to Nuclear Waste Project Office, Carson City, NV, 1993).

26. R. L. Folk, *Geograph. Physique Quat.*, Ottawa, in press (1994).
27. S. D. Malinin, *Physical Chemistry of Hydrothermal Systems with Carbon Dioxide* (Nouka, Moscow, 1979).
28. C. A. Hill, *Special Report no. 6* (Technology Resource Assessment Corp. Rept. to Nuclear Waste Project Office, Carson City, NV, 1993).
29. Y. V. Dublyansky, *Quarterly Report no. 3* (Technology Resource Assessment Corp. Rept. to Nuclear Waste Project Office, Carson City, NV, 1994).
30. Y.-G. Liu and R. A. Schmitt, *Progress Report-June* (Technology Resource Assessment Corp. Rept. to Nuclear Waste Project Office, Carson City, NV, 1994).
31. J. S. Stuckless, Z. E. Peterman, J. F. Whelan, D. R. Muhs, *Geol. Soc. Am. Abstr. Prog.*, 23, A118 (1991).
32. R. W. Spengler and Z. E. Peterman, *Proc. Int. High Level Radioactive Waste Conf.*, April 28-May 3 (Am. Soc. Civil Engineers-Am. Nuclear Soc., Las Vegas, NV, 1991), p. 1416-1422.
33. B. D. Marshall, Z. E. Peterman, J. E. Stuckless, *Proc. Int. High Level Radioactive Waste Management Conf.*, April 26-30 (Am. Soc. Civil Engineers-Am. Nuclear Soc., Las Vegas, NV, 1993), p. 1948-1951.
34. Z. E. Peterman, R. W. Spengler, F. R. Singer, R. P. Dickerson, *ibid*, p. 1940-1947.
35. Z. E. Peterman and J. S. Stuckless, *ibid*, p. 1559-1566.
36. For a complete data chart, map of all collection sites, and source references refer to C. A. Hill and C. M. Schluter, *Data Chart, Yucca Mountain and Vicinity: updated June, 1994* (Technology Resource Assessment Corp. Rept. to Nuclear Waste Project Office, Carson City, NV, 1994).
37. B. D. Marshall and S. A. Mahan, *Proc. Int. High Level Radioactive Waste Conf.*, May 23-27 (Am. Soc. Civil Engineers - Am. Nuclear Soc., Las Vegas, NV, 1994), p. 2685-2691.
38. Z. E. Peterman *et al.*, *ibid*, p. 1316-1323.

39. T. M. Johnson and D. J. DePaolo, *Report LBL-33000, UC-403* (Lawrence Berkeley Lab, Univ. California, Berkeley, CA, 1993), p. 18-20.
40. B. J. Szabo and T. K. Kyser, *Geol. Soc. Am. Bull.*, **102**, 1714 (1990).
41. J. F. Whelan and J. S. Stuckless, *Proc. High Level Radioactive Waste Management Conf.* (Am. Nuclear Soc., La Grange Park, IL, 1992), p. 1572-1581.
42. J. M. Sowers, *et al.*, in *This Extended Land*, D. L. Weide and M. L. Faber, Eds. (Geol. Soc. Am., Cordilleran Sect. Mtg., Las Vegas, NV, 1988), p. 137-157.
43. R. E. Zartman and L. M. Kwak, *Geol. Soc. Am. Abstr. Prog.*, **23**, A117 (1991).
44. R. E. Zartman and L. M. Kwak, *Proc. Int. High Level Radioactive Waste Management Conf.* (Am. Soc. Civil Engineers-Am. Nuclear Soc., Las Vegas, NV, 1993), p. 1953-1959.
45. J. B. Paces, E. M. Taylor, C. Bush, *Proc. Int. High Level Radioactive Waste Conf.*, May 23-27 (Am. Soc. Civil Engineers-Am. Nuclear Soc., Las Vegas, NV, 1994), p. 1573-1580.
46. R. S. Harmon, *Quarterly Report no. 6* (Technology Resource Assessment Corp. Rept. to Nuclear Waste Project Office, Carson City, NV, 1993).
47. I. J. Winograd, B. J. Szabo, T. B. Coplen, A. C. Riggs, *Science*, **242**, 1275 (1988).
48. I. J. Winograd *et al.*, *Science*, **258**, 255 (1992).
49. T. B. Coplen, I. J. Winograd, J. M. Landwehr, A. C. Riggs, *Science*, **263**, 361 (1994).
50. D. L. Bish and J. L. Aronson, *Clays and Clay Minerals*, **41**, 148 (1993).
51. Strontium isotope analyses were made by thermal ionization mass spectrometry in the laboratories of S. Moorbath, Oxford University, UK and of X. Palacz, University of California, Santa Cruz, USA; carbon-oxygen isotope analyses were made by gas-source mass spectrometry in the stable isotope laboratories of J. Hoefs, University of Göttingen, FRG and A. Campbell, New Mexico Tech, USA; U-series dates were made by alpha spectrometry in the laboratory of D. Ford, McMaster University, Canada; fluid inclusion studies were conducted by D. Alderton, University of London, UK and Y. Dubylansky,

McMaster University, Canada; Instrumental Neutron Activation Analyses were performed by Y.-G. Liu and R. Schmitt, Oregon State University, USA; scanning electron microscopy, X-ray diffraction analyses, cathodoluminescence, and petrographic analyses were performed by H. Monger, New Mexico State University, USA; and trace element analyses were performed by Chemex Laboratory, Reno, NV, USA. Drafting was by D. Lopez. Special thanks to D. Livingston for field support and overall encouragement. Funding for this project was received from the Nuclear Waste Project Office, State of Nevada.

TABLE AND FIGURE CAPTIONS

Table 1. Summary of field, petrologic, and petrographic features of controversial calcite/opal, groundwater spring, and true pedogenic deposits, Yucca Mountain.

COVER. Controversial calcite/opal vein and slope calcrete along a segment of the Paintbrush fault, west Busted Butte, Yucca Mountain. Photo: D. E. Livingston.

Figure 1. Map showing major faults, locations mentioned in the text, and the extent of the proposed waste repository (circle). A = Bare Mountain fault, B = Windy Wash fault, C = Solitario Canyon fault, D = Sundance fault, E = Ghost Dance fault, F = Bow Ridge fault, G = Paintbrush Canyon fault, H = Stagecoach Road fault, BMMD = Bare Mountain Mining District, WMD = Wahmonie Mining District, BB = Busted Butte, BC = Black Cone, BMH = Black Marble Hill, CS = Cane Springs, DE = Diatomaceous Earth ("Horsetooth") site, DH = Devils Hole, ET = Eleana trench, LC = Little Cone, LWC = Lathrop Wells Cone, NT = New Trench, OHS = Oasis Hot Springs, PAF = Pull Apart fault, RC = Red Cone, 106 = Site 106, 199 = Site 199, T8 = Trench 8, T14 = Trench 14, WM = Wahmonie Mound, WT-7 = WT-7 well pad, WW = Wailing Wall.

Figure 2. Calcite/opal (white material) directly along fault, Wailing Wall. Away from the fault the mineralization dies out. The fault is part of the Stagecoach Road fault system and is recognizable by slickensides and offset beds. Photo: C. A. Hill.

Figure 3. Composite photo of dissected sand ramp, west Busted Butte, showing calcite/opal vein along the fault (a) and at an angle from fault (b_1, b_2), and slope calcrete which emanates from the vein and continues down gradient to the toe of slope and beyond (c). The highest slope calcrete is at b_2 , which position corresponds to the b_1-b_2 vein. It is important that such calcite/opal slope calcrete is not found on sand ramps uncut by faults (e.g., on some parts of east Busted Butte). If this slope calcrete is pedogenic in origin, it should occur everywhere on the sand ramps and not just along, or downslope from, faults. Photo composite: C. A. Hill.

Figure 4. Splayed-vein and slope calcretes along a segment of the Paintbrush Canyon fault, west Busted Butte, Yucca Mountain. Photo: D. E. Livingston.

Figure 5. A TPD about 1 m thick, Fortymile Wash-Midway Valley. A pedogenic calcrete horizon this thick would have taken hundreds of thousands (or more) years to have formed. This TPD is not located along a fault but is laterally extensive across the valley. Photo: C. A. Hill.

Figure 6. (A) Photomicrograph of fine-grained CCOD, Trench 14, showing the absence of a clastic silicate-grain framework. (B) Photomicrograph of framework-grain supported fabric with each grain coated by calcite, from a modern calcic (Bk) soil horizon on a slope west of Busted Butte. Crossed polars (x 100). Photomicrographs: H. C. Monger.

Figure 7. (A) Pods and seams of pure-textured, pearly opal (a,b,c) in a matrix of dense, buff-colored, mixed-textured calcite/opal (d). Lighter-colored sections are very soft and porous (easily scratched), massive-textured calcite/opal (e). Note the holes (vesicular/phenocystic texture) throughout the mass, especially in the dense, buff-colored calcite/opal (f,g), but also in the massive-textured calcite/opal (h). Also note how the vesicles seem to line up in bands (i and elsewhere). (B) Using a UVG-54

Mineralight, this photo (same position as A) illustrates bands of pure, uraniferous opal fluorescing a brilliant green (a,b,c) in a mixed-textured calcite/opal matrix which does not fluoresce (d,e,h). Sample is from Trench 14. Photos: C. M. Schluter.

Figure 8. (A) Laminated texture where the individual layers are a few millimeters thick.

Note the dark reaction rim at the edge of the Tiva Canyon Tuff where it comes in contact with the calcite/opal matrix. Under thin section this rim does not appear to have been altered or invaded by the calcite/opal; rather, it appears to be a "baked" rim possibly caused by hot solutions. Sample is from WT-7. (B) Two banded samples from the Wailing Wall, showing bands (a few centimeters thick) of alternating mixed, massive, or powdery texture. Photos: C. M. Schluter.

Figure 9. Vesicular/phenocystic texture. (A) Note how the vesicles are aligned in rows along roughly-layered banded sequences; lighter bands (a), darker bands (b), or along wavy flow texture (c,d). This sample was collected along a fault in the Bare Mountain Mining District west of Yucca Mountain and shows that the calcite/opal textural types are regional features. (B) Note how the vesicles occur in both the mixed-textured, buff-colored calcite/opal (a), and also across the boundary into the powdery-textured matrix (b). This sample was collected from Trench 14 and may indicate that the mixed and powdery texture formed penecontemporaneously, with degassing of solutions creating the vesicular texture. Photos: C. M. Schluter.

Figure 10. Two examples of invasive texture: (A) where dense, buff-colored, calcite/opal of mixed texture (a) has "invaded" a powdery-textured mass composed primarily of calcite (b), Wailing Wall; (B) where a "blob" displaying powdery texture (a) has "invaded" a calcite/opal banded mass of mixed texture, WT-7 (b). Photos: C. M. Schluter.

Figure 11. Root-cast texture from west Busted Butte where the CCOD often consist of a tangled mass of root casts. The root casts are large in diameter suggesting that trees or

large plants with deep roots once grew on the slope, such as occur today at the modern Cane Springs (CS; Fig. 1). Lens cap for scale. Photo: C. A. Hill.

Figure 12. Ratios of elemental abundances in representative samples of CCOD (A), GSD (B), and TPD (C) to nearby A-horizon soils and normalized to the thorium abundances in the samples and soils. The INAA (Instrumental Neutron Activation Analyses) procedure is the same as reported by (30) and references therein. The Fortymile Wash TPD (FMW 1,3) were not used in the comparisons because they contain a high amount of detrital matter ("soil matrix"). Since only two TPD were used for comparisons (RC, RVF) the results must be considered tentative.

Figure 13. $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic data for all reported surface and subsurface, rock, water, calcite, CCOD, GSD, and TPD, Yucca Mountain (36). The double XX represents averages for all the samples including the low- ^{87}Sr data points (e.g., X = 45, Trench 14) or for all the samples minus the low- ^{87}Sr data points (e.g., X = 43, Trench 14). The Bare Mountain range of values represents altered and unaltered, undifferentiated Late Proterozoic and Paleozoic rocks (38).

Figure 14. $^{87}\text{Sr}/^{86}\text{Sr}$ values (open rectangles) for CVD plotted as a function of distance from the modern water table (0 m). The range of $^{87}\text{Sr}/^{86}\text{Sr}$ values for CCOD is shown by the horizontal arrows in the lower part of the figure. The vertical arrow marks the average $^{87}\text{Sr}/^{86}\text{Sr}$ value for ground water in the Cenozoic volcanic aquifer. Present-day $^{87}\text{Sr}/^{86}\text{Sr}$ values for the volcanic rocks are shown by the filled rectangles. Rock units are: TS = Topopah Spring; CH = Calico Hills rhyolite; PP = Prow Pass; B = Bullfrog; T = Tram; dl = dacite lava; LR = Lithic Ridge; A, B, C = unnamed units. After (20).

Figure 15. Carbon-oxygen diagram (PDB) for all reported vein and slope CCOD at Yucca Mountain and vicinity (36).

Figure 16. Carbon-oxygen diagram (PDB) for all reported CVD; A = saturated zone, B = shallow phreatic zone, C = unsaturated zone (36). CCOD (D) are represented in both Figs. 15 and 16 by the ellipse labelled "controversial calcite/opal." Asterisks (*) denote

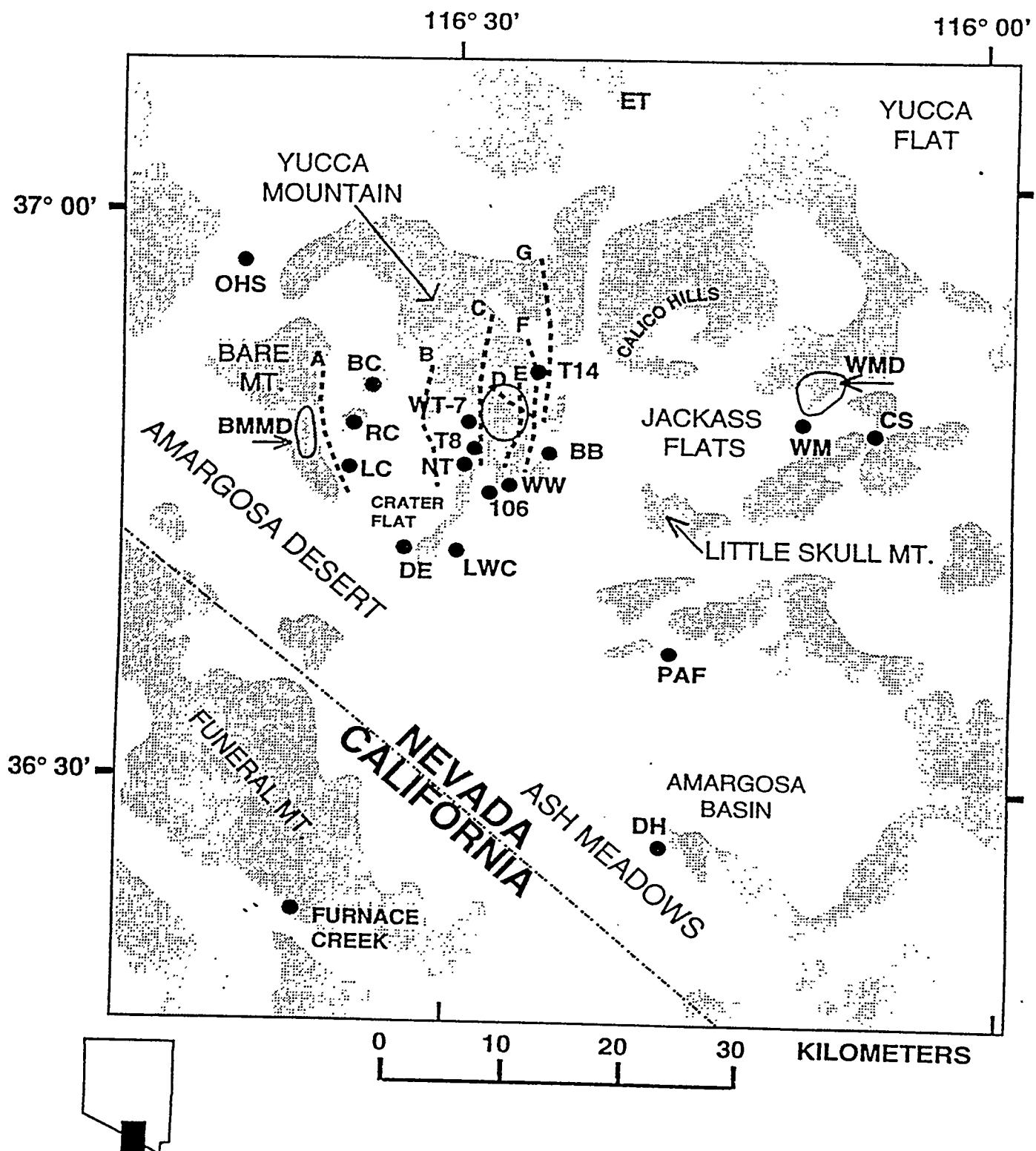
carbon-oxygen values for TPD; collection site elevations are in meters. Altered limestone = LS and unaltered limestone = hatched area.

Figure 17. Trench 14 and Busted Butte CCOD and Devils Hole, Amargosa Basin, Furnace Creek Wash, and Crater Flat GSD plotted on a U-series isochron diagram (36). "Pedogenic" carbonate field is from (45) but may be CCOD affected by open-system conditions rather than being representative of TPD. Asterisks (*) represent unleached (or relatively unleached) samples from Trench 14 and Busted Butte (46).

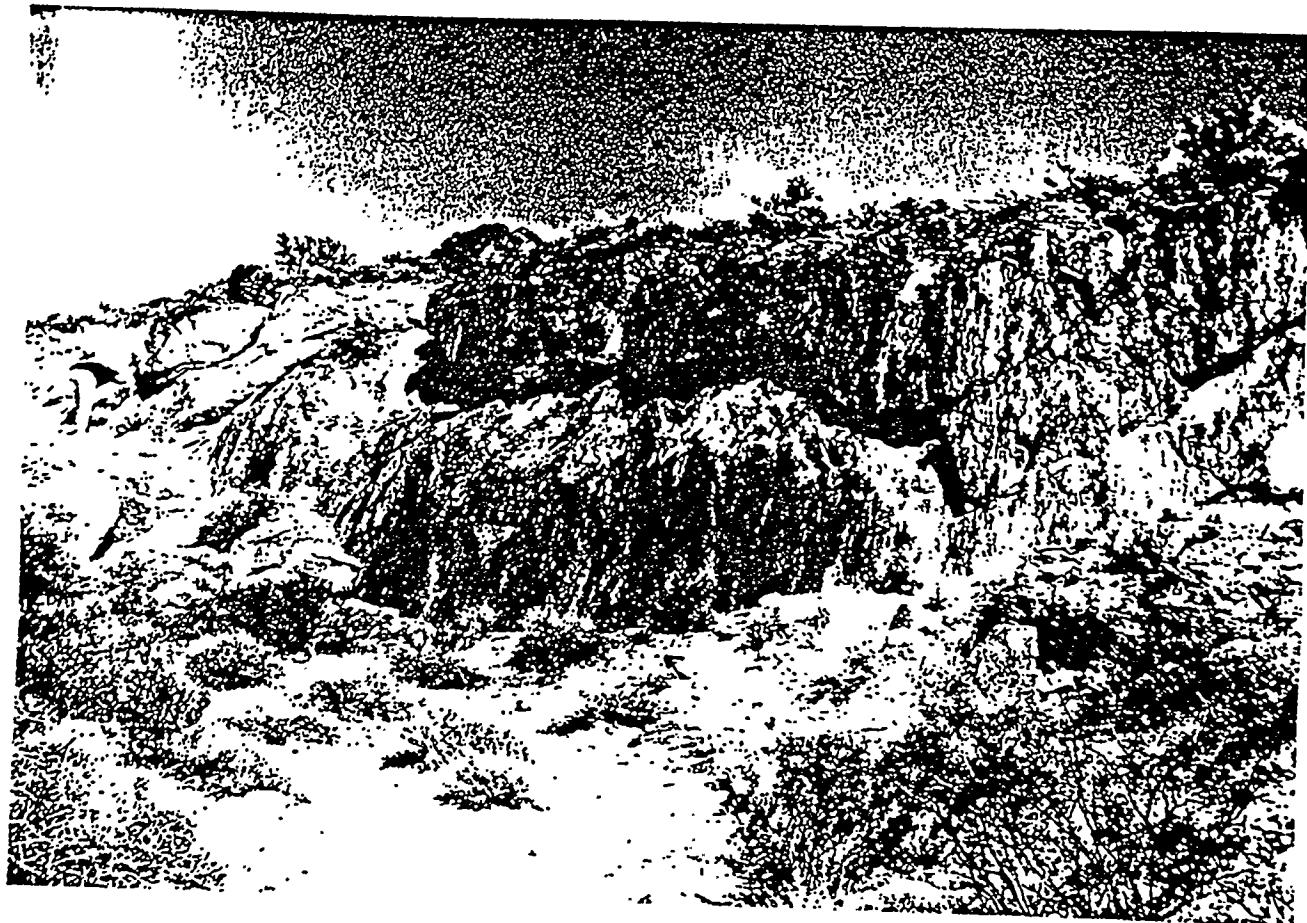
Figure 18. Age of CCOD, GSD (springs, S), and CVD (drill holes, DH) (36). Note that the ages of these deposits seem to cluster into discrete groups.

Table 1 Summary of field, petrologic, and petrographic features of controversial calcite/opal, groundwater spring, and true pedogenic deposits, Yucca Mountain

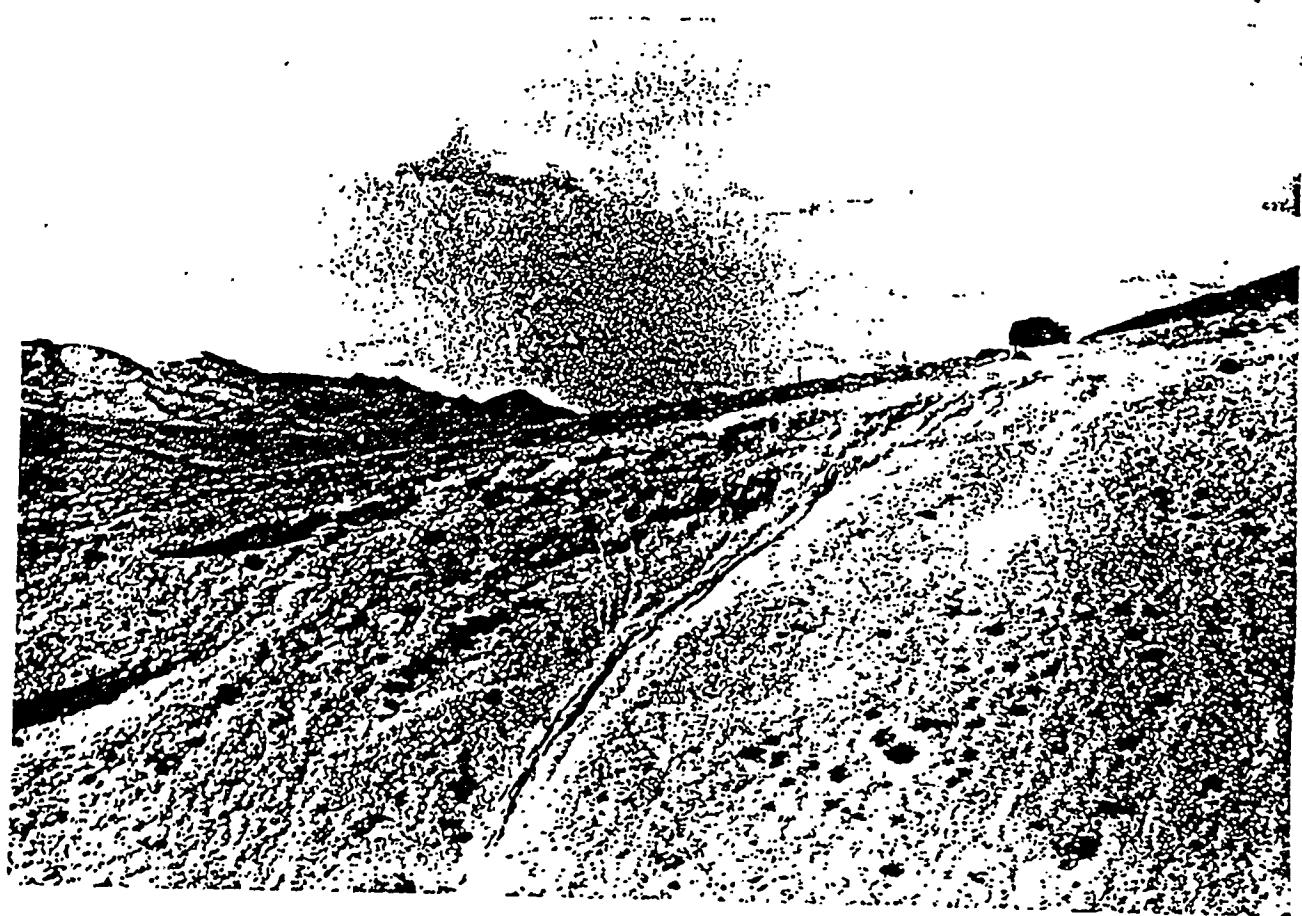
	<u>Controversial calcite/opal</u>	<u>Groundwater Spring</u>	<u>True Pedogenic</u>
<u>Example locations</u>	Trench 14, Busted Butte, Pull Apart fault, WT-7, Wailing Wall, Bare Mtn.	Site 199, Diatomaceous Earth, Wahmonie Mound, Cane Spring	Red Cone, Pull Apart fault, Fortymile Wash
<u>Field occurrence</u>	Vein and slope calcretes associated with faults	Mounds at former spring sites	Laterally extensive horizons; carbonate build-up within older geomorphic surfaces
<u>Hand specimen</u>	Variety of massive and laminar features	Massive	Morphogenetic sequences of (21)
Petrologic textures	Pure, mixed, massive, banded/laminated, flow, brecciated, patchy, ooidal, vesicular/phenocrystic, root-cast, invasive, speleothemic, veined	Powdery	Powdery, ooidal
Fluorescence	Pure opal bands fluoresce brilliant green	non-fluorescent	mostly non-fluorescent tiny stringers of opal fluoresce, RC, FMW
Phosphorescence	Yellow-green-blue glow with excitation flash (WT-7, T14, WW), $T_{\text{fm}} = <60^{\circ}\text{C}$ (29)	-	-
<u>Microscopic features</u>			
Crystal size	Micrite and spar, very fine-grained, 1 μm to 200 μm	Micrite and spar, 1 μm to 70 μm	Micrite to spar, <1 μm to 2mm
Sorting	Variable: well to poor	Usually well sorted	Usually poorly sorted
Shape	Anhedral to euhedral	Subhedral to euhedral	Anhedral to euhedral
Mineralogy	Calcite, opal; minor sepiolite, quartz; trace pyrite/chalcopyrite	Calcite (>95%), 199 Opal (>95%), DE Calcite-gypsum, WM Opal (common), CS	Mainly calcite; detrital quartz and feldspar; opal stringers, void linings, root structure and ooid fillings, Non-luminescent
Cathdoluminescence	Non-luminescent	Non-luminescent to slightly-luminescent	Non-luminescent
Alizarine Red stain	Red	Red	Red
K-ferricyanide stain	negative	generally negative	negative
Organic matter	Microbial structures, large root casts to small rhizoliths	Diatoms (199, DE, CS) Ostracods (199) No microbes (WM) No rhizoliths observed	Microbial structures, small rhizoliths

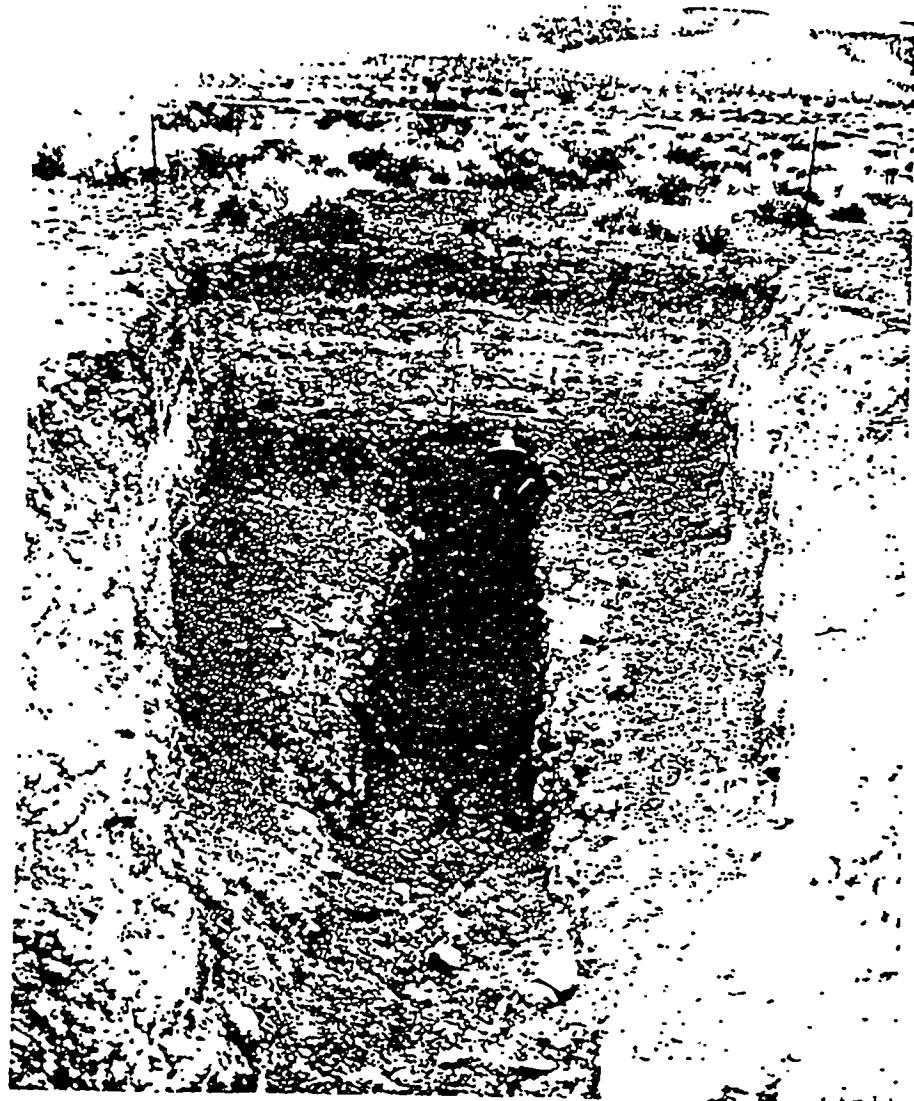


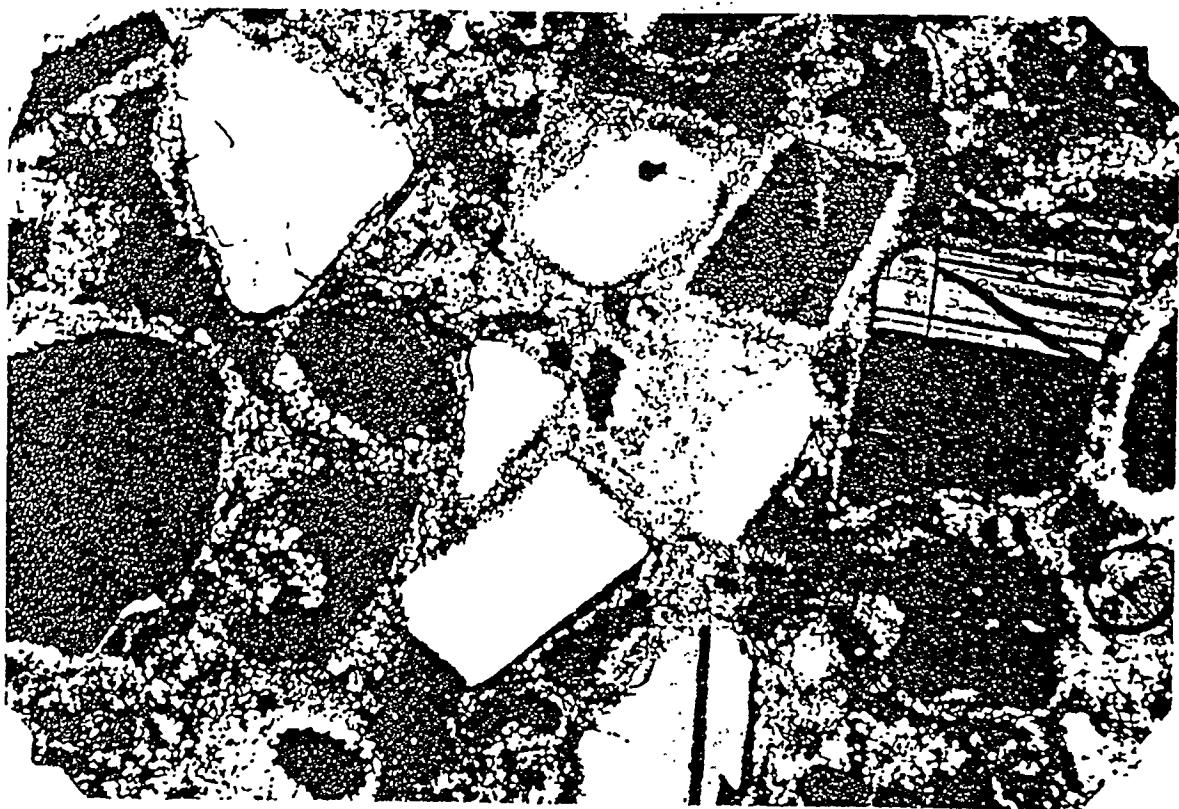
Hill, Fig. 2



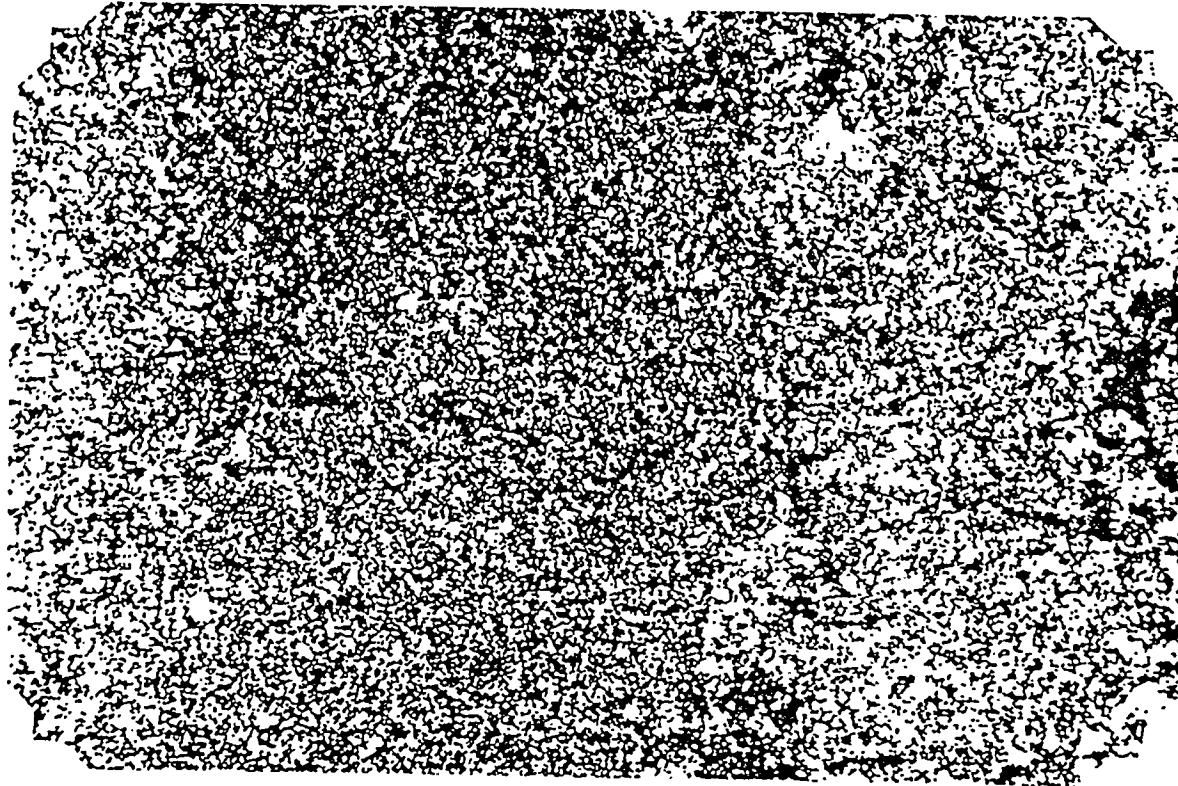






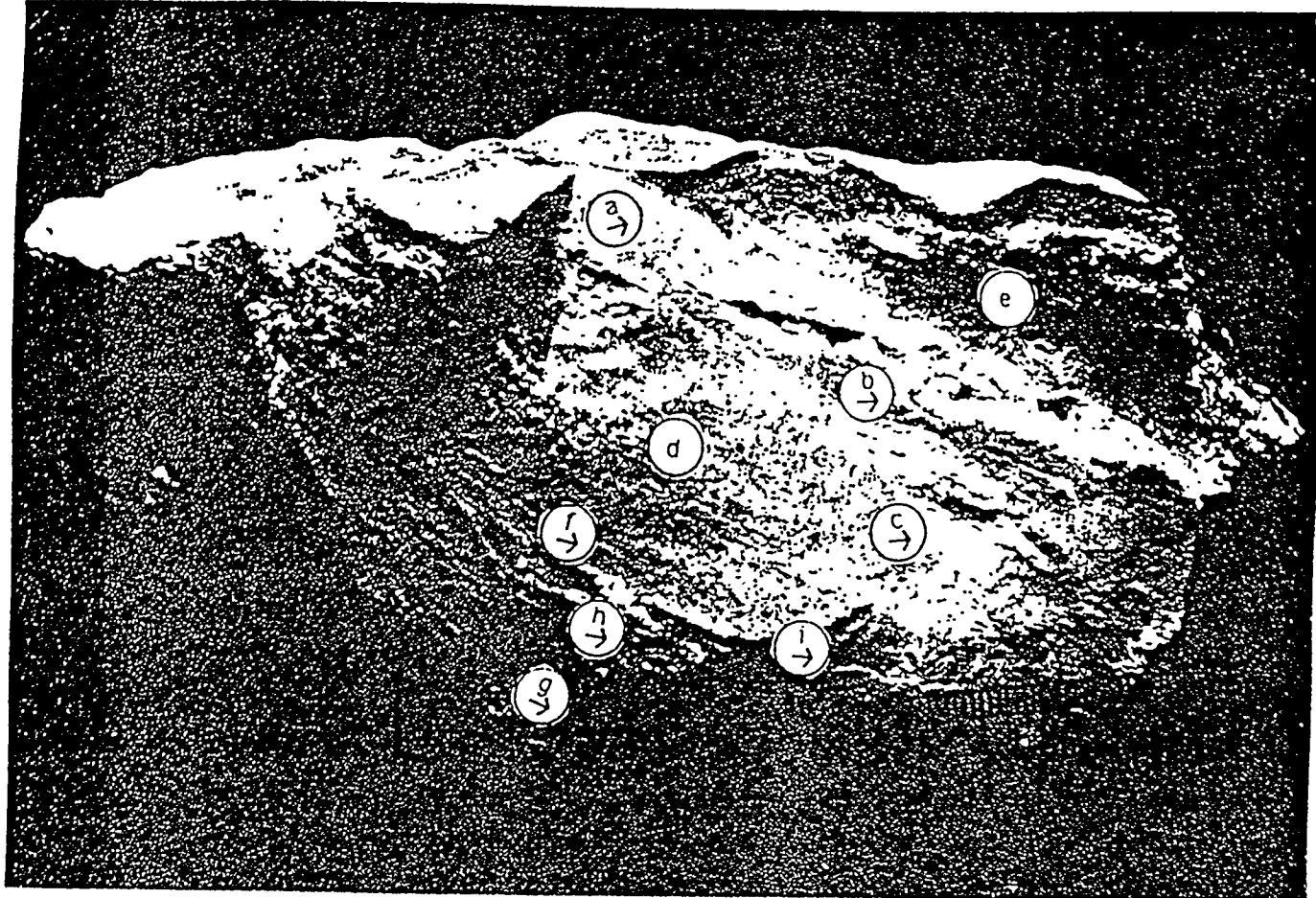


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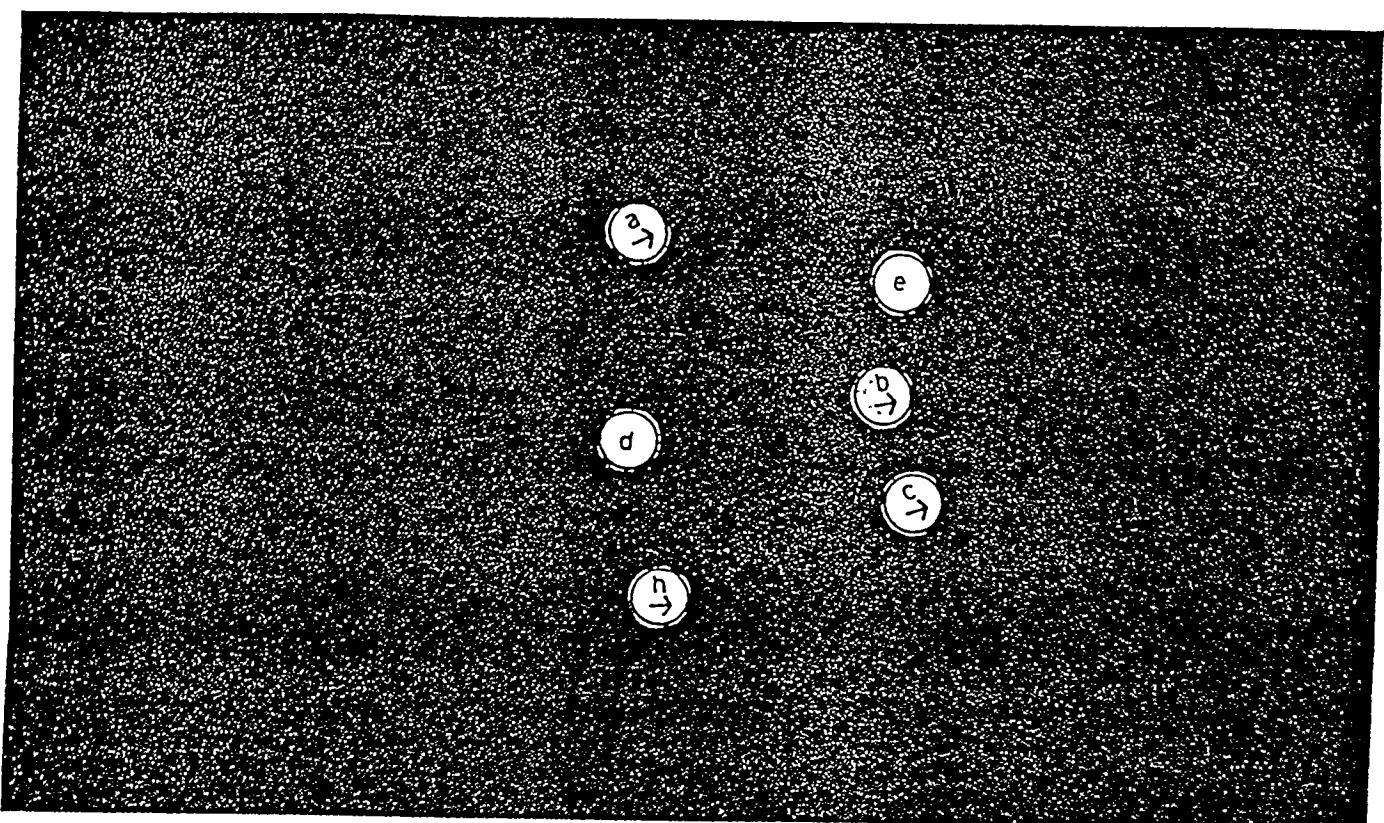


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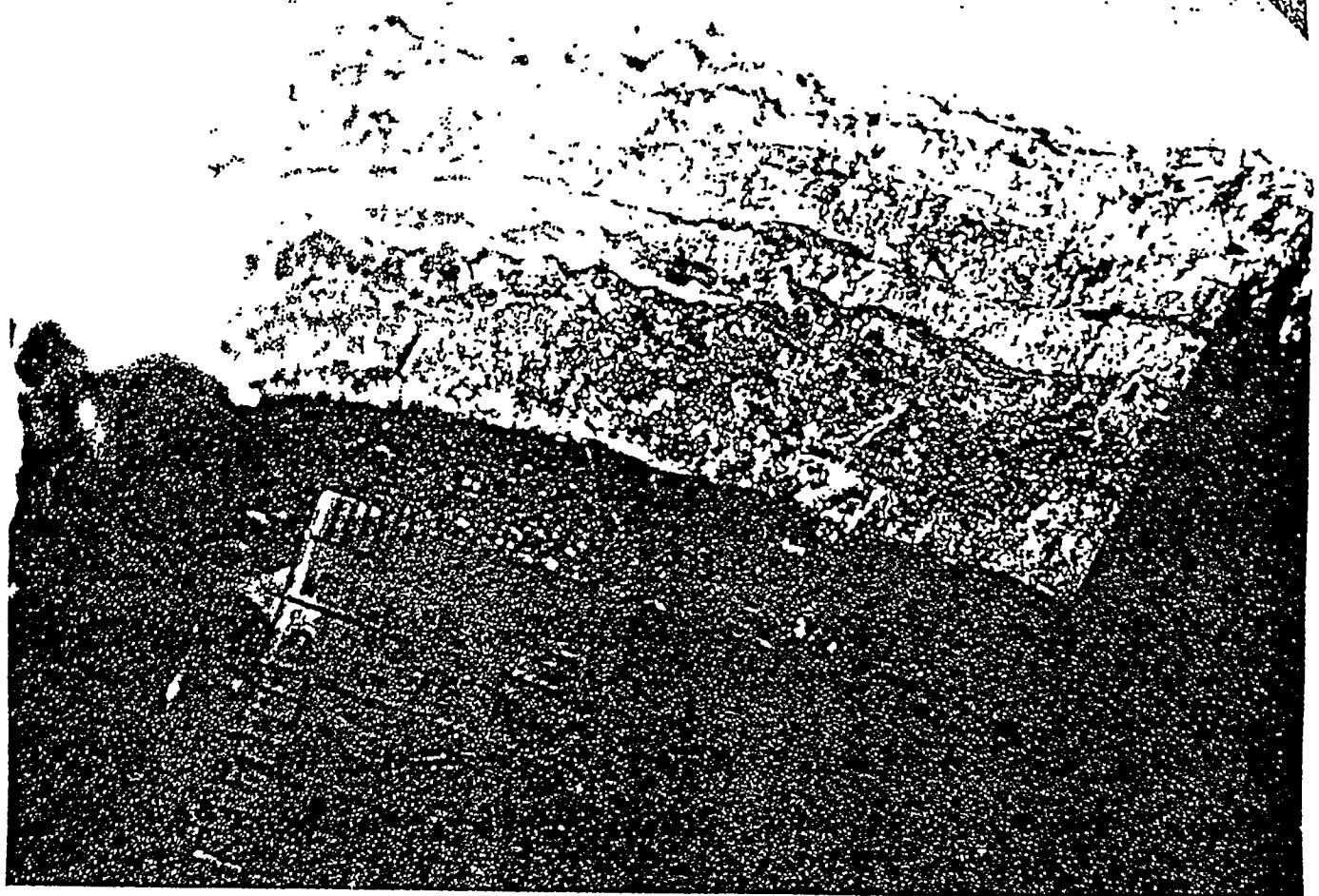
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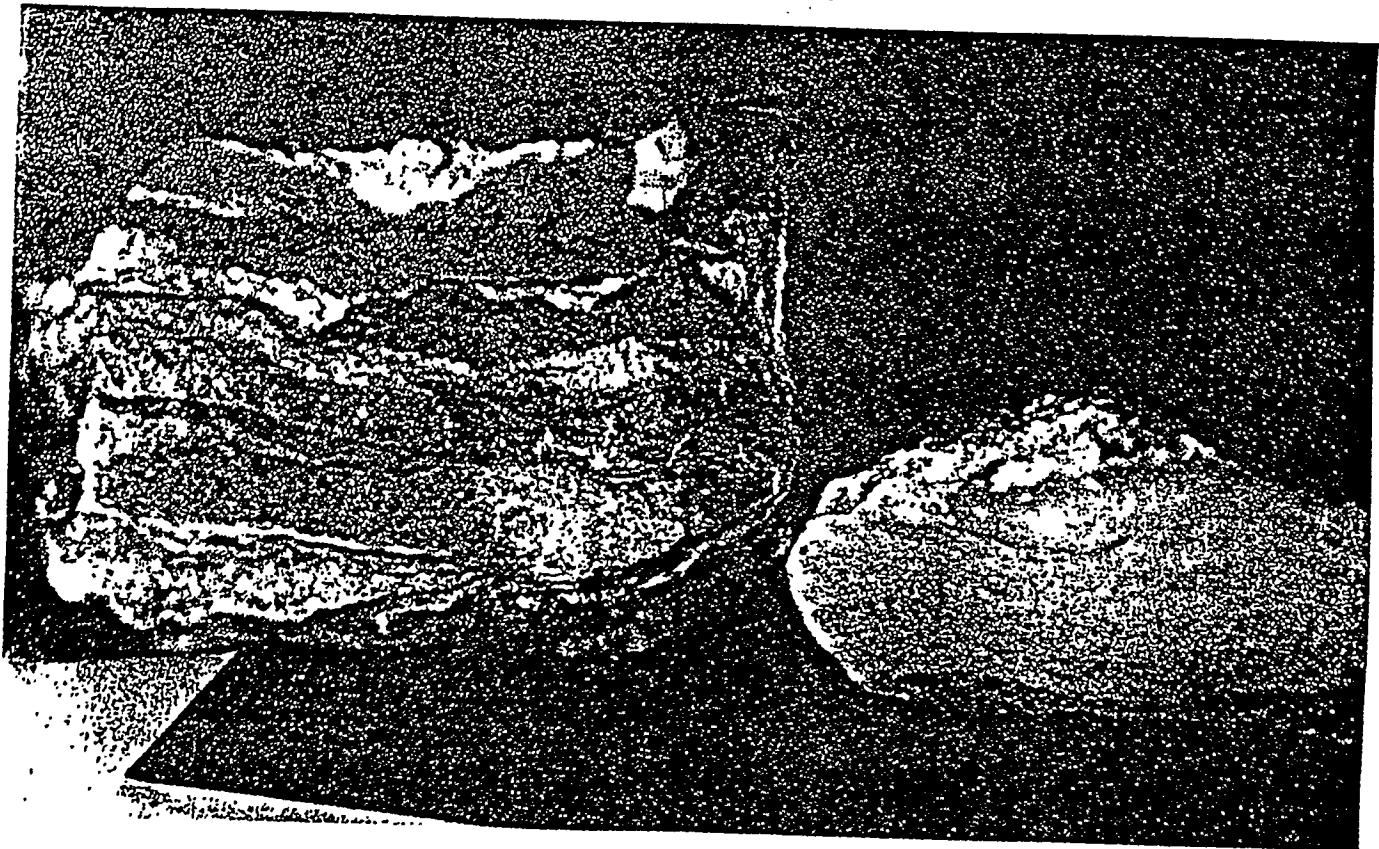
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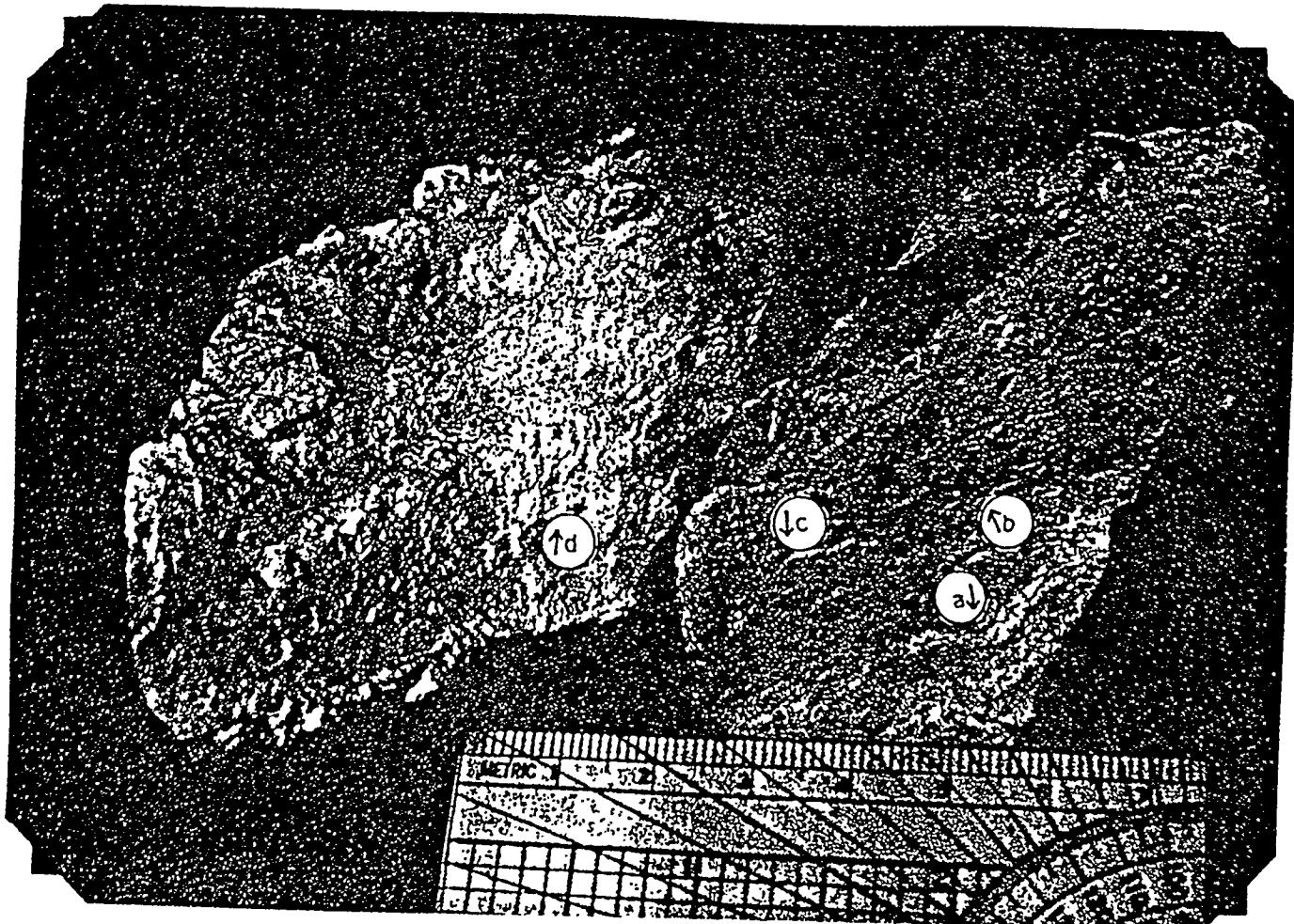
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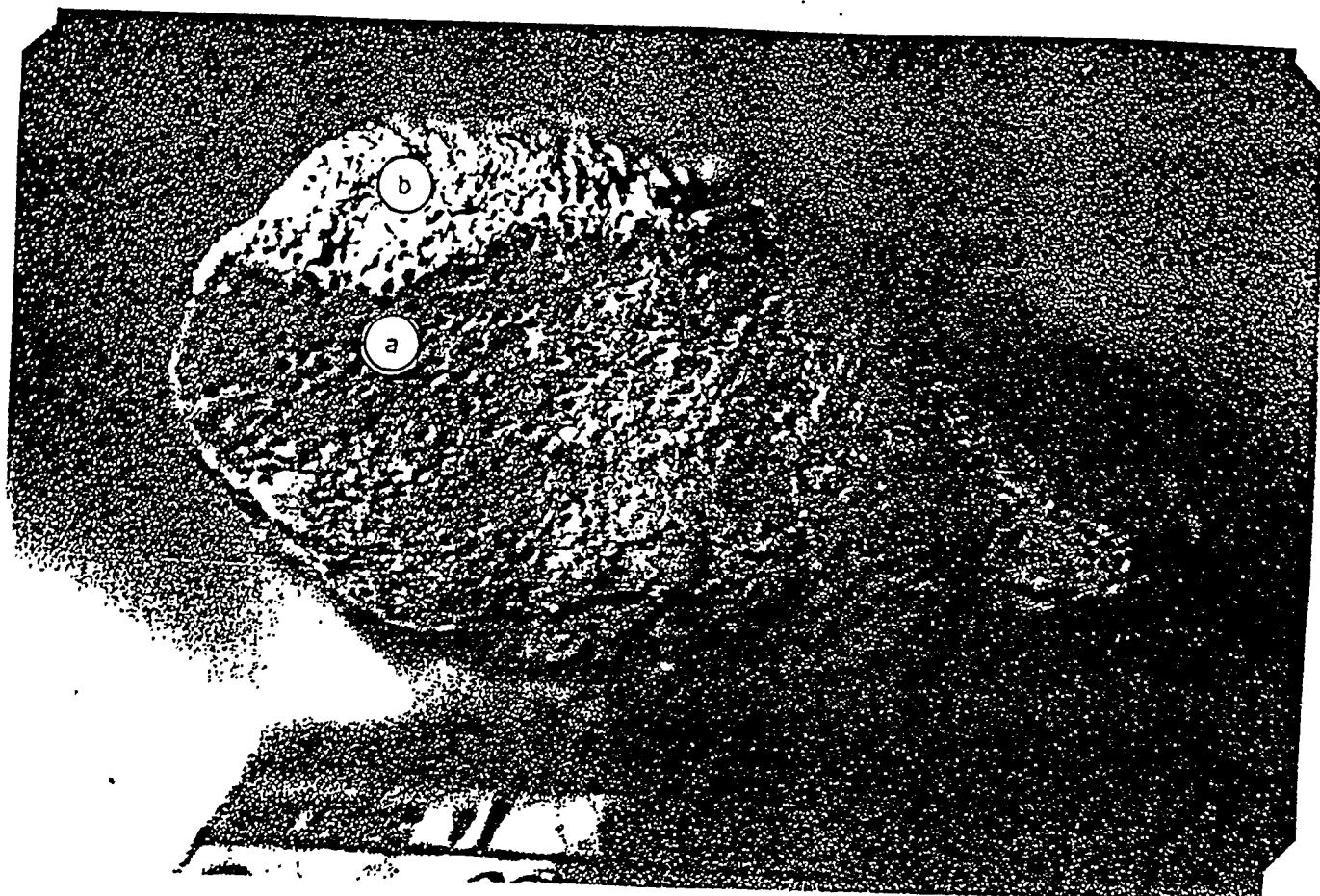
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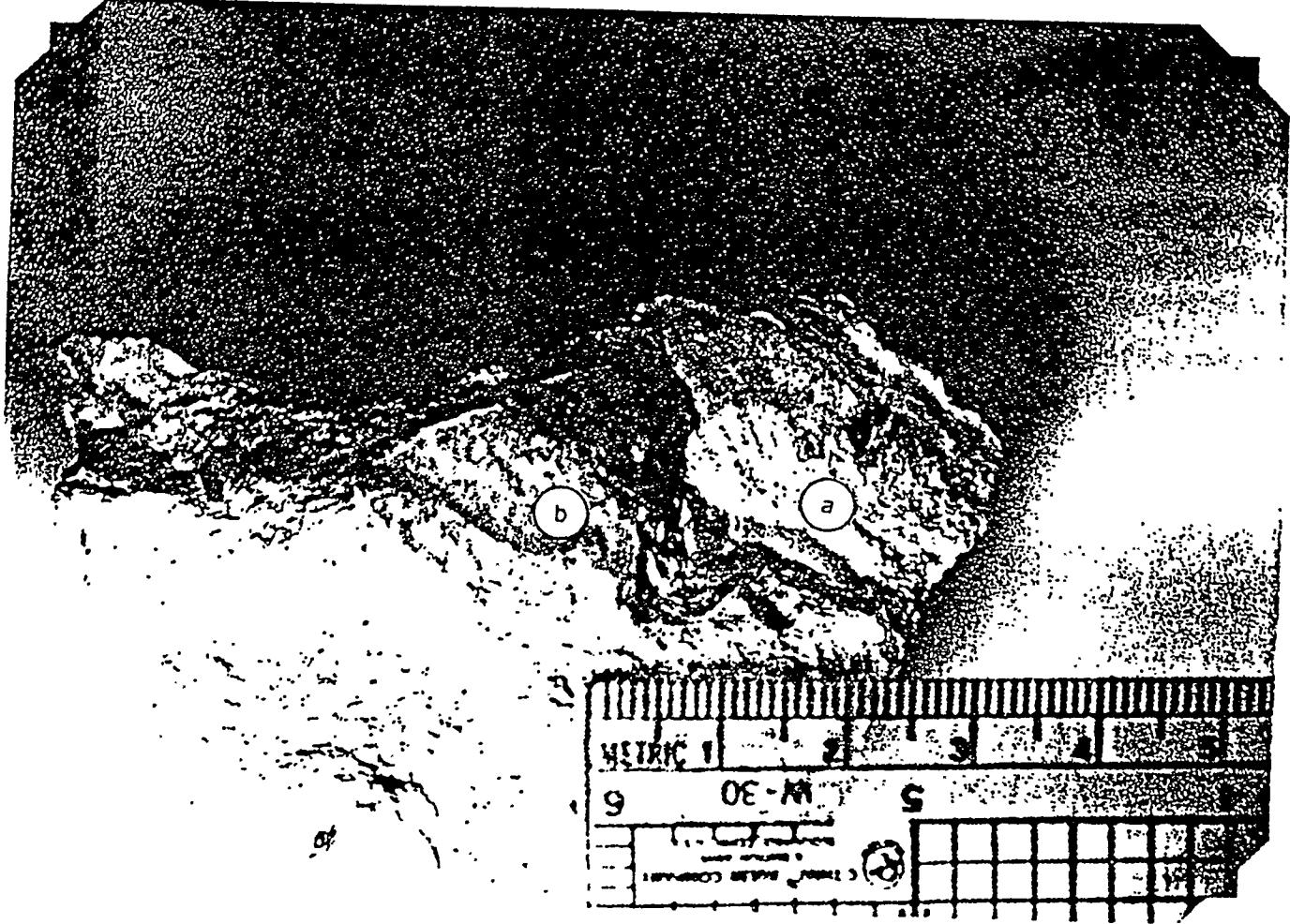
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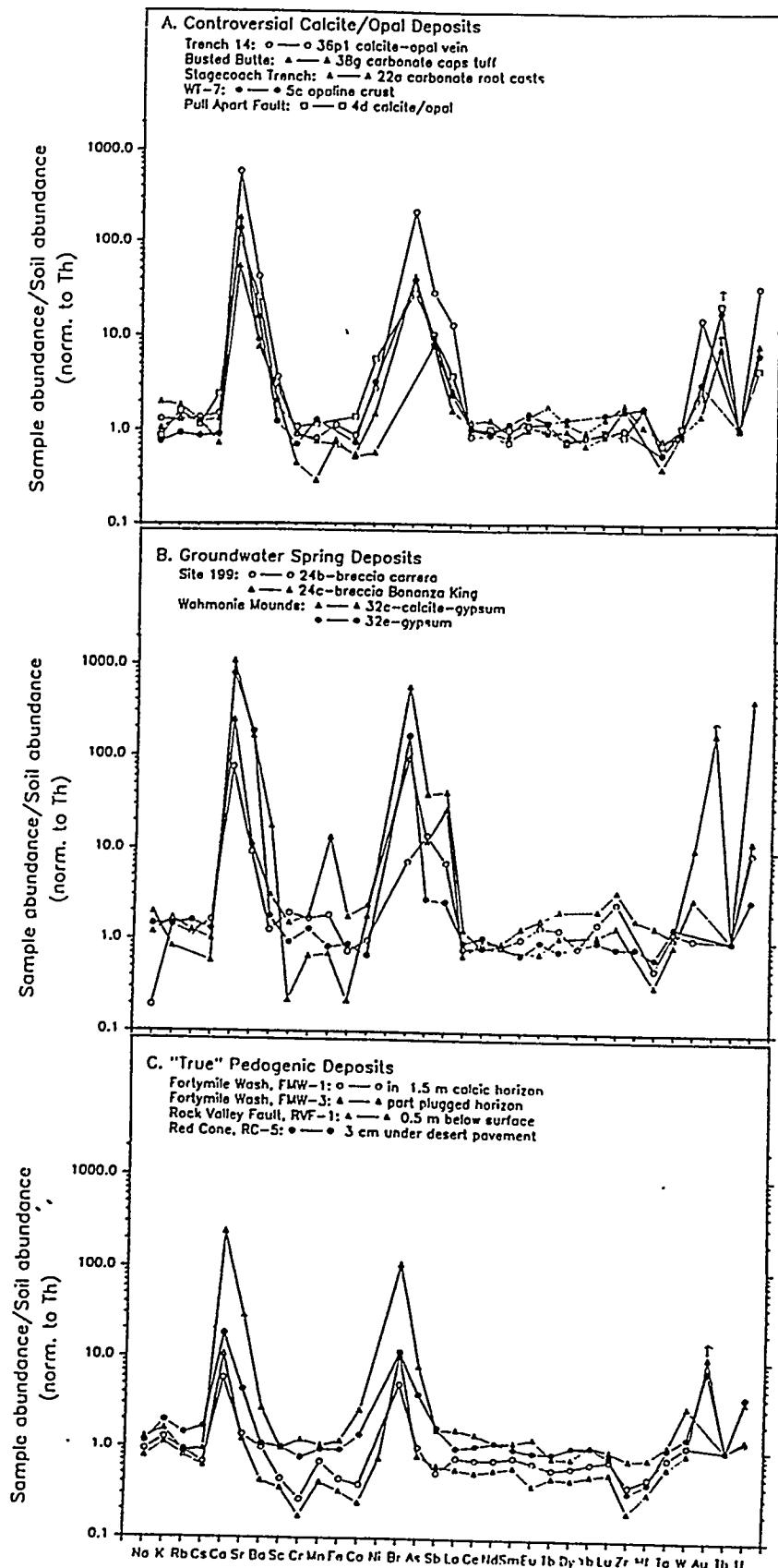
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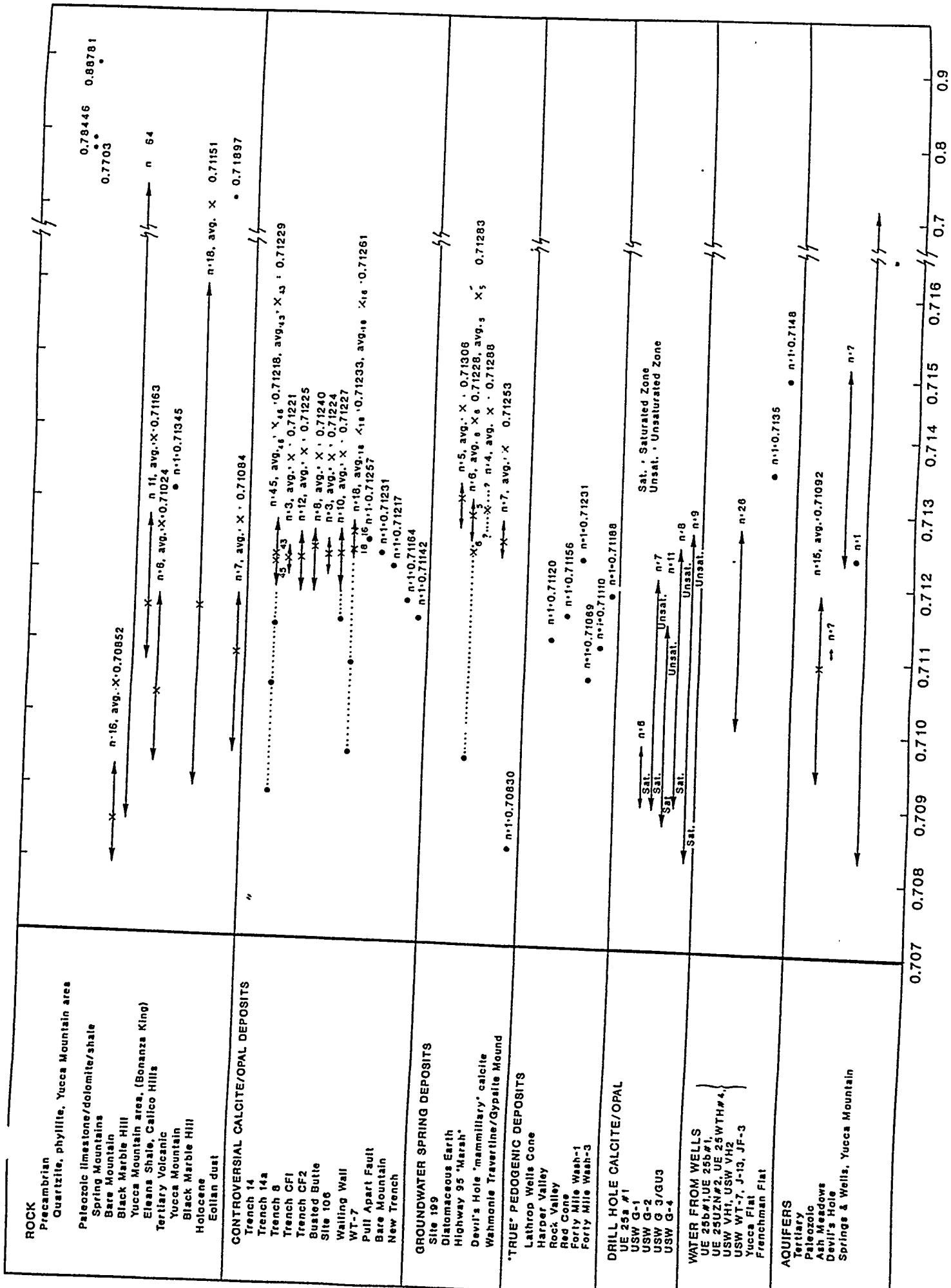


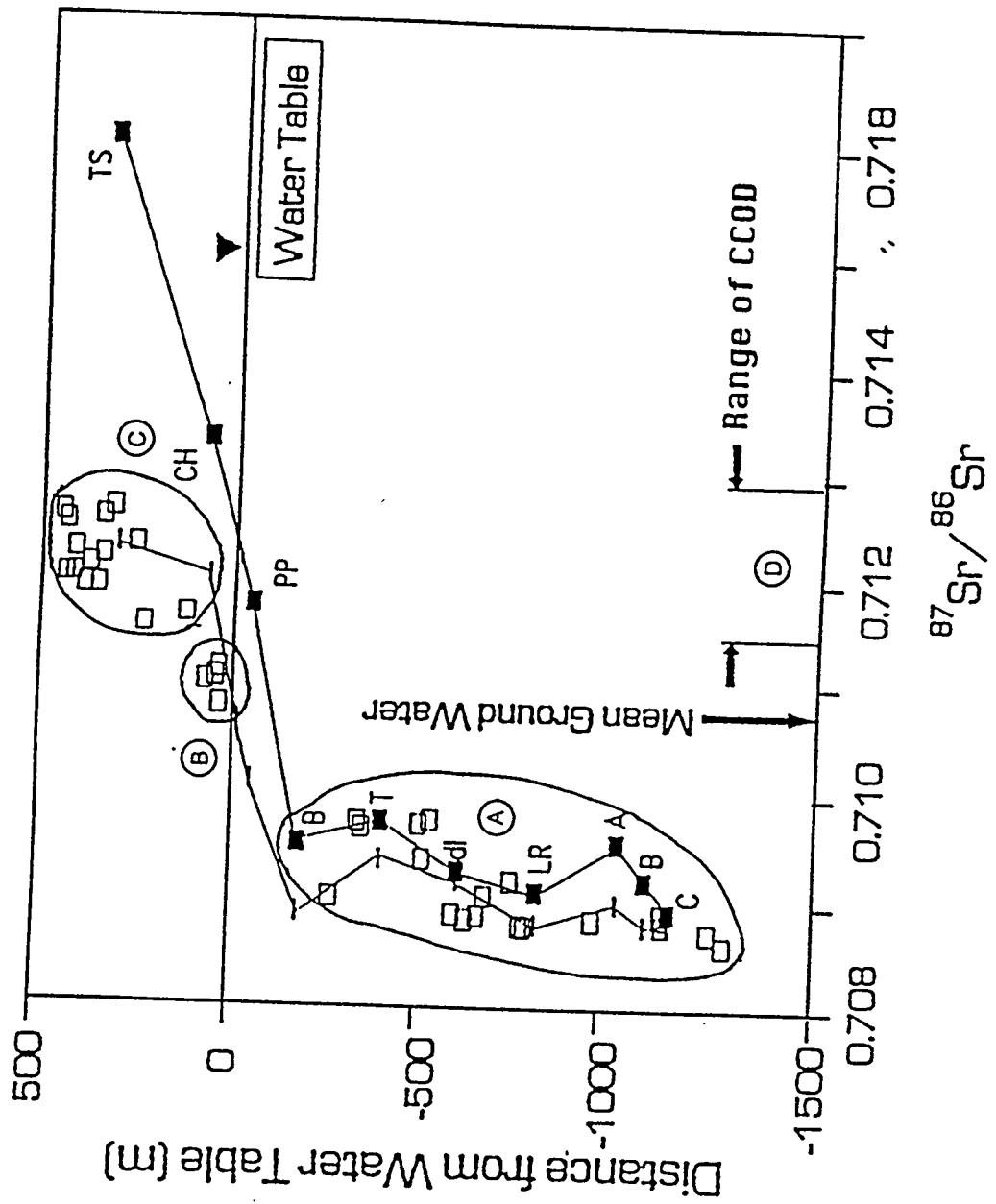
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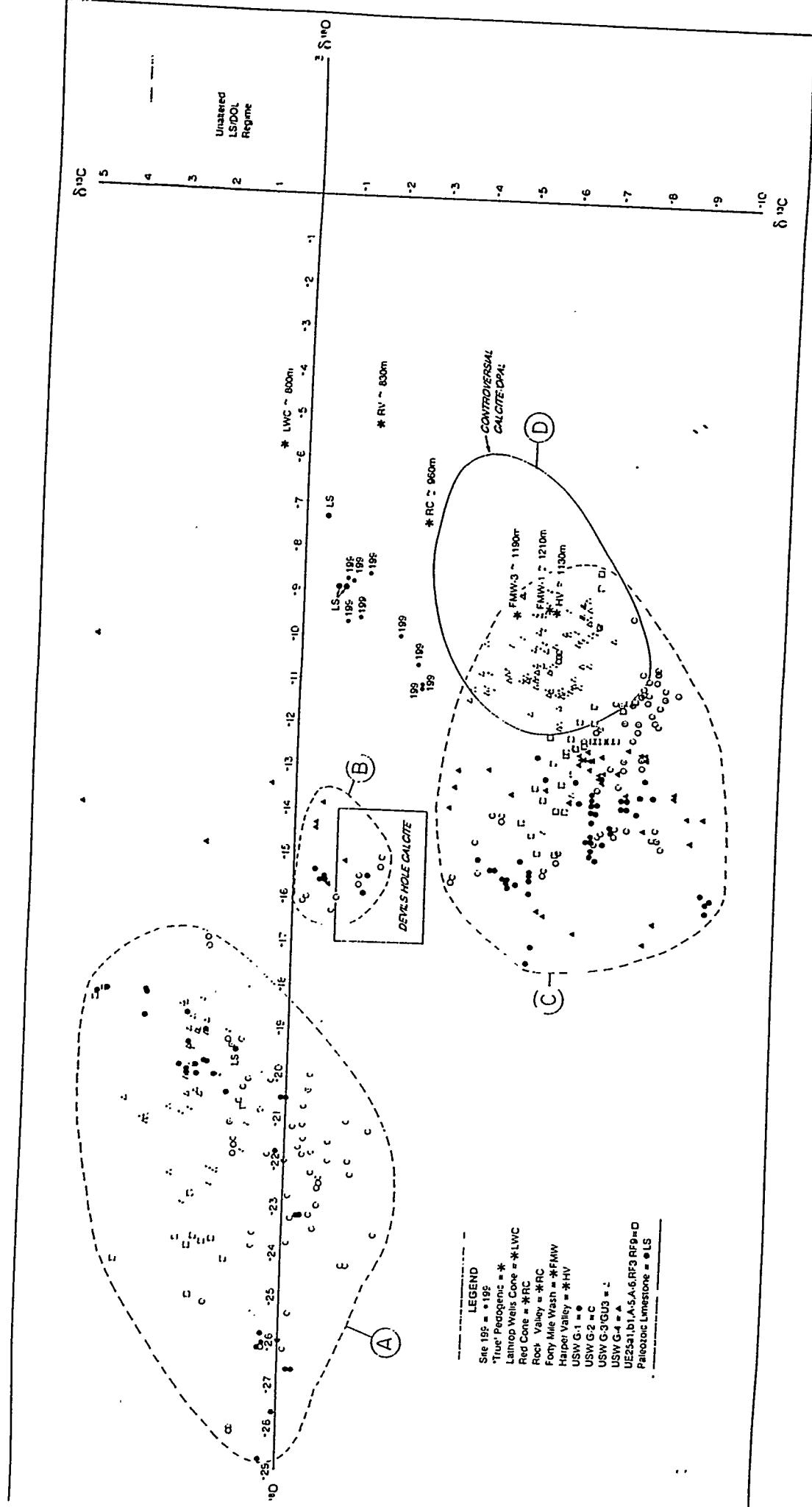


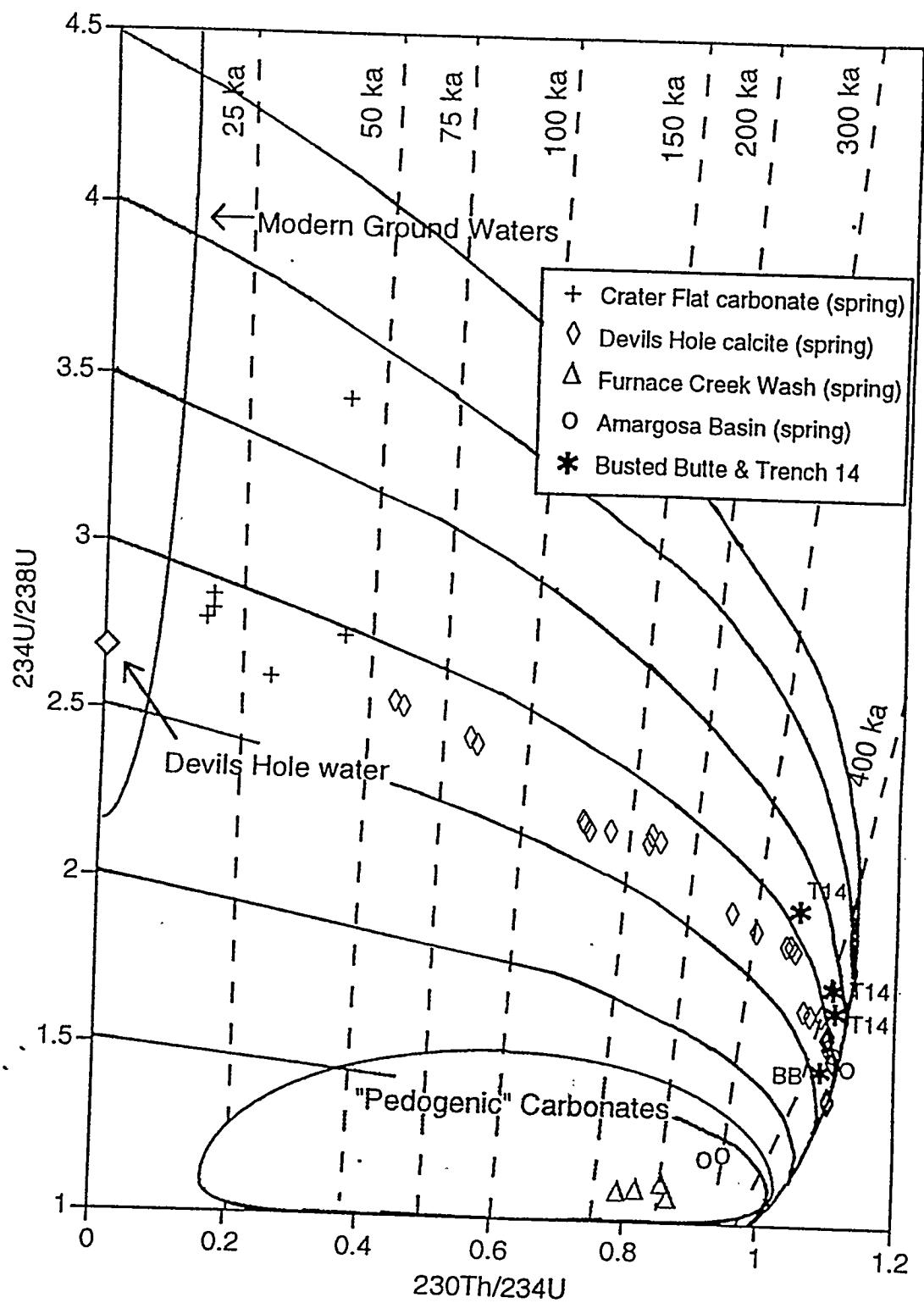












ATTACHMENT 3

PART 1

**Review of the paper: "Fluid Inclusion Studies of
Calcite Veins from Yucca Mountain, Nevada, Tuffs:
Environment of Formation" by Y. V. Dublyansky,
Academy of Sciences of Russia**

Review of the paper:

**Fluid Inclusion Studies of Calcite Veins from
Yucca Mountain, Nevada, Tuffs: Environment of Formation**

by Edwin Roedder, Joseph F.Whelan and David T.Vaniman

Introduction

The author's understanding of genesis of Yucca Mountain calcite is given as the following: "The unsaturated zone calcite appears to have precipitated from rainwater that have descended along interconnected fractures carrying dissolved carbonate from the overlying soil environment; CO₂ escape from the fluid is a likely mechanism for the precipitation of this calcite within the unsaturated zone tuffs" (p.1854). The paper is an attempt to prove this model.

The text contains many solid theoretical arguments pertaining to fluid inclusions and various methods of their study, as well as detailed descriptions of methods used. The information on obtained results, however, is amazingly meager. For instance: about 35 % of the text's length are dedicated to theoretical and methodic aspects of thermometry by fluid inclusions, but no single datum on homogenization temperature is given.

Moreover, some data given by the authors are in contradiction with their own model. At least two lines of evidence: textures of calcite and fluid inclusions do not favor "descending" model, so the later doesn't look convincing. In the following text I shell argue my opinion.

I. Textural features of Yucca Mountain calcite

The authors describe textures encountered in unsaturated zone below 15 m as the following: "...calcite is sparry and locally coats fractures and forms drusy masses within litophysal cavities". This calcite is further interpreted as a film water precipitate. However, sparry and drusy textures are not typical of film precipitates. The environment where calcite deposited from gravitation-driven water films most often found (and thus, studied in detail) in the caves of vadose zone. The speleothemic coatings formed from water films have normally mamillary laminated textures. The thicknesses of individual laminas are rarely exceed 1 mm. Sparry and drusy textures are virtually unknown in vadose (subaerial) cave environment (except some specific and rare cases like drip-splash crystals on tops of stalagmites or some aerosol formations). Instead, such textures rather suggestive of phreatic (subaqueous) environment.

In saturated zone: "...calcite occurs as veins, often with chlorite and quartz or chalcedony, and as a replacement cement of altered tuffs" (p.1854). Quartz, chalcedony and opal also occur in unsaturated zone of Yucca Mountain. The declaration that calcite in saturated zone "...is part of an alteration mineralogy formed during a low-temperature hydrothermal event that occurred ~10,4 ma..." is not necessarily true. This age was determined by means of K/Ar technique by illite/smectite (Bish & Aronson, 1993). The only date obtained for calcite in saturated zone by U-series is >400 ka (Szabo & Kyser, 1990).

II. Thermometry by fluid inclusions in calcite

Calcite is a mineral rather difficult for fluid inclusion studies. It is very cleavable and susceptible for re-crystallization even at low temperatures. This causes many problems on the stage of sample preparation. Besides, it is extremely difficult to distinguish

primary, pseudo-secondary and secondary character of inclusions on the basis of petrographic observations. The existing criteria for distinguishing the origin of inclusions "...are not absolute, and many are merely suggestive... they must be applied with care and with awareness of the considerable ambiguity that exists" (Roedder, 1984, p.12; emphasis by Roedder). This is especially true when dealing with calcite.

Fluid inclusion is called **primary** if it was trapped during growth of a studied part of a crystal or a mineral body. It is classified as **pseudosecondary** if it was trapped after the studied part of a crystal was crystallized but before the growth of a whole crystal (mineral body) ceased. The inclusion is called **secondary** if it was trapped after the growth of mineral ceased. Primary and pseudosecondary inclusions characterize different temporal stages in evolution of mineral-forming environments, while secondary inclusions are characteristic of post-mineralization environments. Thus, the T_{hom} measured by fluid inclusions may characterize temperatures of fluids in which the crystal studied was bathed during or after its growth.

In case of Yucca Mountain it is unimportant what type of inclusion was used for thermometry: In either case high T_{hom} would mean that the rocks of Yucca Mountain were flooded with thermal water on a certain stage of its history.

To apply this approach, however, we must be sure that inclusions studied (a) were trapped as homogeneous phase, and (b) they were not damaged (shranked or opened and leached) after the entrapment. This is hard to prove if one deal with a single inclusion but much easier when dealing with groups of inclusions. If such a group consist of two-phase inclusions with uniform V/L ratio, it gives us a strong indication that this particular group may provide reliable and meaningful thermometric information. Inclusions in such groups yield normally T_{hom} varying within 1-2 °C interval.

As it is obvious from the citations below, such groups of inclusions have been found in calcite from unsaturated zone in Yucca Mountain: "Suitable primary liquid-filled inclusions, containing a shrinkage vapor bubble, were found in only a few samples" (p.1857) and: "Most of the suitable inclusions occurred in groups, with an apparently uniform and small V/L ratio..." (p.1858). Surprisingly, the authors give us no numerical information on T_{hom} measured on these suitable inclusions (we may deduce that these have been measured from another quotation: "The meager T_h data indicate that at the time of growth of host calcite the ambient temperature was equal to the measured T_h ..." (p.1585)). The authors only mention that "...small V/L ratio... visually indicated that the inclusions had formed at low temperatures, probably $< \sim 100$ °C" (p.1858). Such an approach, i.e., visual estimation instead of instrumental is quite astonishing by itself and I will discuss it latter. But there is something else that can be deduced from this short author's description. The minimal temperatures of entrapment could not have been lower than approx. 40 °C, otherwise the shrinkage bubbles simply would not have nucleated. (The physical mode of a gas bubble nucleation is described by the authors quite thoroughly on p.1858. However, their statement that "Inclusions in the 10-20 micrometer range, particularly if trapped at < 100 °C, almost never nucleate a bubble..." is not correct. My 14-year experience of work with low-temperature hydrothermal calcite shows that inclusions of 5 to 25 μm in size do nucleate bubbles yielding T_{hom} of 100-40 °C).

Thus, even being not provided with numerical data we may conclude that at a certain stage of its history the calcite studied was bathed in (and, quite probably, deposited from) fluids of approx. 40 to 100 °C. Obviously, these temperatures at depth -130-314 m can not be attributed to the "normal" geothermal gradient of ~ 34 °C/km (Sass e.a., 1980) in unsaturated zone.

Visual method of thermometry by fluid inclusions

Visual method has been designed by Nikolay Yermakov (1944). The *rationale* of this method was given by the author as the following: "...the homogenization method requires a special apparatus for heating minerals to high temperatures under the microscope. This is not always possible at any given place and time. We attempted therefore to produce empirical curves for the most common vein minerals that would help an investigator, equipped with any kind of microscope, in drawing tentative conclusion in regards to the temperature minima at which a given hydrothermal mineral may have been formed" (Yermakov, 1965, p.108), and: "The curves we have derived are suited only for approximate determination of the anticipated homogenization temperatures of inclusions. One may resort to them only in the absence of microthermochamber (thermostage, YuD)" (*Ibid*, p.116).

Author's interpretation of data

I have shown that (1) the authors were aware of the presence of two-phase fluid inclusions in calcite from unsaturated zone of Yucca Mountain, and (2) these inclusions imply temperatures higher than those that could have been induced by normal geothermal gradient. How are these data interpreted in the authors model developed in final section "ENVIRONMENT OF FORMATION OF THE CALCITE VEINS"? They are simply ignored. The authors even deny their own previous statements implying "...the absence of two-phase, liquid + vapor inclusions in the upper thousand feet of the USW G-1 borehole..." (p.1859; *cf.* with quotations above). This statement is also in contradiction with data on T_{hom} measured in unsaturated zone calcite from the same drill hole: 81 °C at -221 m (YMP, 1993), and other drill holes of the area: 78 to 227 °C in USW G-2 and G-3 (Bish, 1989 and Bish & Aronson, 1993). Obviously, these data can not be explained satisfactory within the model of deposition of calcite from thin films of rainwater seeping through the unsaturated zone.

III. Gases in inclusions

Crushing technique

The method of formation temperature estimation according to amount of immersion oil entering the inclusion on crushing seems to be very hard to calibrate. There are many variables that may play a role in the behaviour of an inclusion.

1. Solubility and diffusion. If we deal with a gas phase consisted of a mixture of non-condensable gases like CO₂, O₂, N₂ and CH₄ we should be aware that (a) every gas is soluble to some extent in the crushing oil and (b) the gases may migrate from gas bubble into the oil and from oil into the bubble due to diffusion. Normally we do not know: - the real composition and partial pressures in a gas phase; - the solubility of these gases in the oil; - the amount of gases already dissolved in the oil (that influence the concentration gradients between the gas phase and crushing oil and control the rate of diffusion); and - the diffusion coefficients for our gases in this particular crushing oil. The diffusion coefficients are often extremely temperature-sensitive (for instance, for glycerol $D \sim \ln(\ln T)$, where D - diffusion coefficient, and T - temperature). Thus, the slightest change in ambient temperature may influence the result.

2. Gas bubble size. Detailed studies of the dynamics of gas bubble dissolution on crushing were performed in Laboratory of fluid inclusions of Institute of Mineralogy and Petrography, Nòvosibirsk in 80-ies. It was found that this process is non-linear and may be subdivided on three stages: (1) very fast but exponentially slowing decrease of a bubble during first 1-2 seconds due to both solution and diffusion; (2) slow decrease; and (3) exponentially accelerating decrease and collapse of a bubble when it reaches a certain "critical" size due to increase of internal pressure. Empirically, the "collapse diameter" was found to be approx. 3-5 mcm. It means that small bubbles (with original diameter <6-8 mcm) may collapse immediately after crushing even if they contain essentially "insoluble" gases.

3. Geometry of inclusions. Fluid inclusions are three-dimensional vacuoles often with irregular shape. So, any estimations of volume phase relationships may not be too accurate.

Thus, the method suggested by the authors is not a "straight forward" one, and the possibility of obtaining reliable data this way is somewhat doubtful.

Mass-spectrometric data

Qualitative mass-spectrometric analysis has revealed N₂, O₂, CO₂, and CH₄ (methane is referred as "major" in the abstract; p.1854). Most unfortunately, no data on sample preparation, method of gas extraction and laboratory procedures have been given. Thus, there is no possibility to assess the reliability of the data. One should be aware, however, about some general problems with gas analyses by destructive methods:

(1) The gases are extracted from a certain amount of calcite. It means that these gases derived from different assemblages of inclusions, that represent different stages of mineral growth, and may even be trapped after the growth ceased (secondary inclusions). There is no way to estimate more or less accurately the ratio of these gases in a final mixture which is analyzed by mass-spectrometry or gas chromatography.

(2) Both most common methods of release of a gas for analysis - decrepitation and crushing - cause significant change in component ratio due to gas reactions at high temperatures and adsorption on mineral surfaces freshly created by crushing.

(3) The gases in calcite may reside not only in inclusions, but also in lattice defects and, most important, they may be adsorbed on clay particles trapped as solid inclusions (these particles have extremely high specific surface). Greyish-white and light-brown color of Yucca Mountain calcite (Whelan e.a., 1994) might indicate the

presence of such clay contaminants. Our studies have shown that the amount of gases recovered by gas chromatography may differ by one to two orders of magnitude for zones with and without clay admixtures in a single calcite crystal (Dublyansky, 1990). Also, at least a part of methane might be related to organic impurities, reported for this calcite (Whelan e.a., 1994). In general, trace amounts of methane are common in low-temperature hydrothermal calcite from elsewhere (Dublyansky, 1990; Dublyansky & Reutski, 1993).

It is a sensible approach, thus, to keep in mind the opinion of the senior author of the reviewed paper given in his excellent treatise earlier: "Although it might seem extreme, I believe that the possibilities of major errors in inclusion analyses are sufficiently numerous that one should simply discount all analytical reports that do not give details on sample size, and the selection, cleaning, and extraction procedures used, as well as the usual statements of analytical methods, sensitivity, accuracy, precision, blanks, standardization, etc." (Roedder, 1984, p.110).

Origin of vapor-rich inclusions

"Vapor-filled inclusions provide, by their very existence, evidence of the presence of a vapor phase along with the liquid phase from which the host crystal grew" (p.1854). This is correct, provided these are undoubtedly primary inclusions. The difficulty of proof of that has already been discussed. So, it would be safer to say that such inclusions evidence the presence of a vapor phase during inclusion entrapment. Several alternative models may be proposed to explain the origin of vapor-rich inclusions. For instance the effervescence of CO₂ is quite typical in ascending hydrothermal solutions due to decrease of both pressure and CO₂ solubility (Malinin, 1979). Exsolving CO₂ (or any other gases) would form vapor bubbles and leave the system. However, if the velocity of upwelling water is high enough, the system would remain heterogeneous and calcite forming would trap vapour-rich inclusions along with liquid-rich ones (possibility 3, discussed by the authors on p.1859). And finally, the

possibility that vapor-rich inclusions were trapped on the latter stages of calcite history during its residence in unsaturated zone should also be considered.

REFERENCES

Bish D.L., 1989, *Evaluation of Past and Future Alterations in Tuff at Yucca Mountain, Nevada, Based on Clay Mineralogy of Drill Cores USW G-1, G-2, and G-3*. Los Alamos National Laboratories, Report LA-10667-MS, 40 pp.

Bish D.L. and Aronson, J.L., 1993, *Paleogeothermal and Paleohydrologic Conditions in Silicic Tuff from Yucca Mountain, Nevada*. Clays and Clay Minerals, v.41, no.2, pp.148-161.

Dublyansky Yu.V., 1990, *Regularities of formation and modelling of hydrothermal karst*. Novosibirsk: Nauka, - 150 p. (In Russian)

Dublyansky Yu.V. and Reutsky V.N., 1993, *Hydrothermal system of some karst massifs of Hungary (by inclusion in minerals)*. Report submitted to Hungarian National Authorities for Nature Conservation, Budapest, 128 p.

Malinin S.D., 1979, *Physical chemistry of hydrothermal systems with carbon dioxyde*. Moscow:Nauka, 111 p.

Roedder E., 1984, *Fluid Inclusions*. Reviews in Mineralogy, v.12. 644 pp.

Sass J.H., Lachenbruch A.H., and Mase C.W., 1980, *Analysis of thermal data from drill holes UE25 a#3 and UE25 a#1, Calico Hills and Yucca Mountain, Nevada Test Site*, USGS Open File Report 80-826, 25 p.

Szabo B.J., and Kysér T.K., 1990, *Ages and stable isotope compositions of secondary calcite and opal in drill cores from Tertiary volcanic rocks of the Yucca Mountain area, Nevada*. GSA Bull, 102:1714-1719.

Yermakov N.P., 1944, *The temperatures of formation of the deposits of optical minerals in Central Asia*. Soviet Geology, 1:34-36 (In Russian)

Yermakov N.P., 1965, *Studies of mineral-forming solutions*. In:D.E. Ingerson, editor. Research on the nature of mineral-forming solutions. International Series of Monographs in Earth Sciences, 22. New York: Pergamon Press. 348 p.

YMP, 1993, *Data released by Yucca Mountain Characterization Project Office.*
December, 1993

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**REVIEW OF THE NAS/NRC REPORT:
"GROUNDWATER AT YUCCA MOUNTAIN:
HOW HIGH CAN IT RISE?"**

**SPECIAL REPORT No. 1
CONTRACT No. 92/94.0004**

**SPECIAL REPORT Submitted to the
Nuclear Waste Project Office
State of Nevada**

December, 1992

Authored by:

C. B. Archambeau

**Review of the NAS/NRC Report:
"Groundwater at Yucca Mountain: How High Can It Rise?"**

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Review of the NAS/NRC Report:
"Groundwater at Yucca Mountain: How High Can It Rise?"
by
Charles B. Archambeau

There are three basic and serious problems that produce disagreement with the conclusions and recommendations of the Academy report. These are: *First, the report ignores a considerable body of critical data relating to the ages and nature of hydrothermal alterations at the site; second, many of the strong conclusions expressed in the report are not reasonably supported by the evidence presented and, in some cases, are inconsistent with data and results available to the committee but which are not cited or used by them; and finally, there are statements describing field relationships and data that are not consistent with the facts or are made in such a way as to be misleading.*

Zircon Age Data: Evidence for Hydrothermal Activity

An example of what can be regarded as a misleading characterization of data is given on page 44 of the report. The Academy Panel states:

"Fission - track dating of eroded fragments of (or detrital) zircons found in carbonate that cements AMC - type fault breccia at Trench 14 and at Busted Butte gives a spread of ages showing heterogeneity of source material, with some zircon ages older and some younger than the age of the bedrock in the immediate region (Levy and Naeser, in press). However, within the analytical uncertainty, most of the ages are about 10-12 Ma, or about the same as those of the dominant volcanic rocks in the region."

However, the Levy and Naeser reference states (p. 17):

"The spread in ages from each sample indicates that there are zircons from multiple sources present. In both samples there are crystals significantly younger and significantly older than the age of the tuff." (Emphasis added.)

In the following paragraph Levy and Naeser go on to show plots of these data and state the basis for their confidence in the observed spread in zircon ages as follows (references quoted are omitted):

"One way to illustrate the spread in the ages is through the use of a probability density distribution plot. The probability density plot sums the normal distribution curves for all the grains in a sample. These curves are calculated from an age and its standard deviation. Figure 6 shows an example of a sample with a single age population; the Fish Canyon Tuff zircons are used as a primary age standard for most fission-track laboratories in the world and the probability curve exhibits a normal distribution. In contrast, samples HD-41-4 and HD-74-2 both show multiple age peaks (Figures 7 and 8). The ages of the individual grains are shown in the histogram beneath the probability curves for all three samples."

The data shown by Levy and Naeser in their Figures 7 and 8 are reproduced in the attached Figure 1. These data clearly show the multiple peaks identified by Levy and Naeser. *Contrary to what is stated by the Panel*, most of the zircon crystals analyzed from each sample show dates considerably less than the Potassium-Argon ages of the host tuff (13 Ma), rather than greater than the age of the tuff. Further, the Panel implies an age for the host tuff of 10-12 Ma, while it is clearly stated to be 13 Ma.

As seriously misrepresentative is the neglect of the Panel to indicate that the authors clearly use the term 'significant' in a technical sense. In fact, the Panel report does not even mention that the authors themselves attach significance to peaks in the distribution and that they do *not*, in any way, suggest that *"within the analytical uncertainty the ages are about the same as those of the dominant volcanic rocks in the region."* This is the Panel's statement, but they do not distinguish this assertion from the previous sentence referencing the paper by Levy and Naeser. They thereby induce the reader to assume that this statement is consistent with the results of the authors. In this way they do not have to explain why their characterization of these data is different from that given by the authors, or even mention that a difference exists.

An examination of the age data, as given in Figure 1, shows that there are ages 4.8 Ma, 6.2 Ma, 7.5 Ma, and 7.7 Ma among the crystals in these two samples. There are

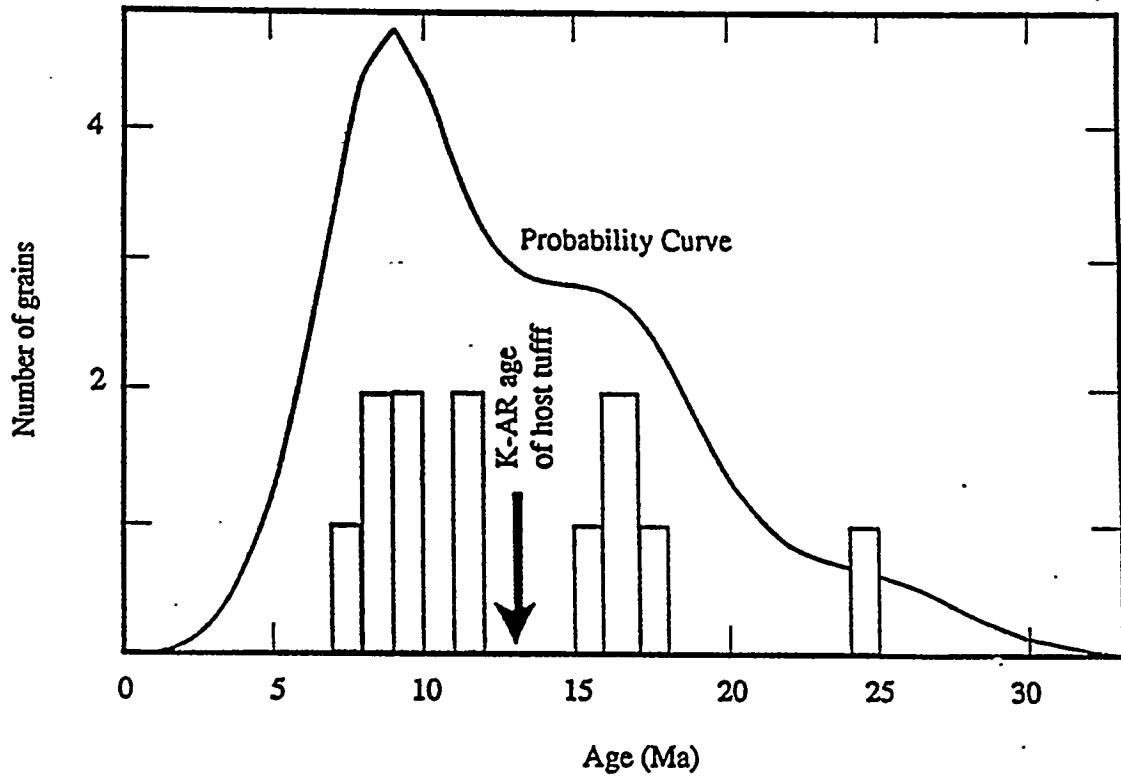
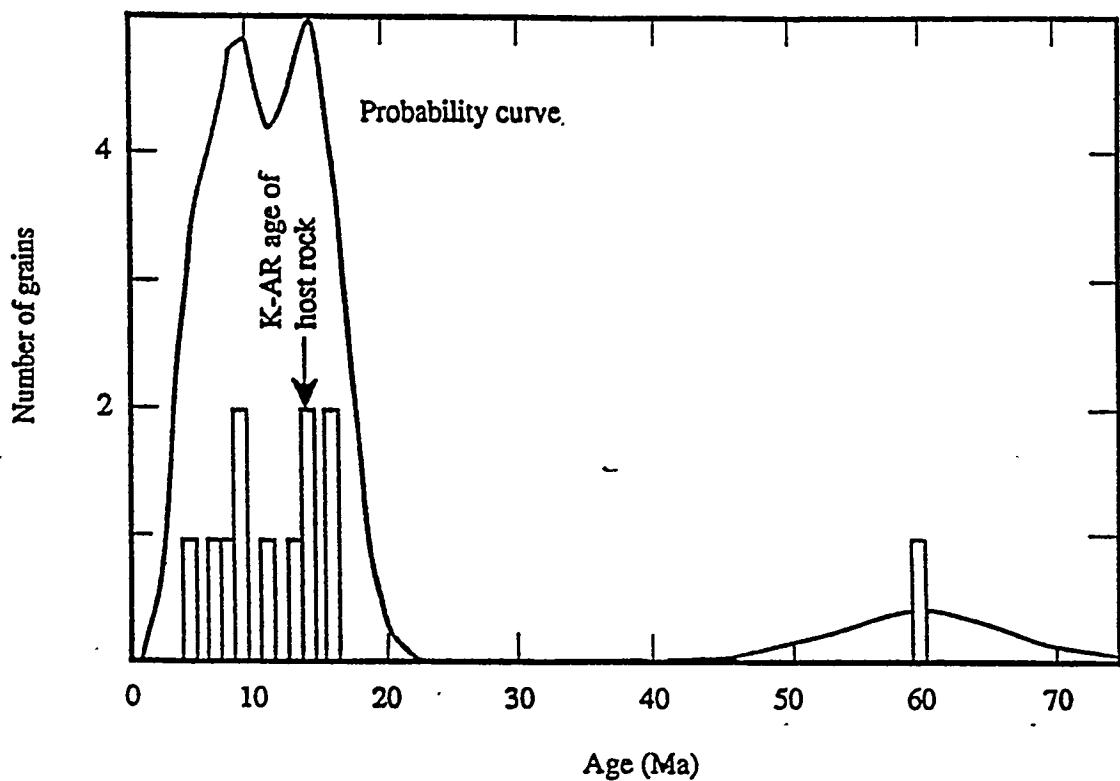


Figure 1. Fission track ages of zircons from breccias at Busted Butte (top) and Trench #14 (bottom). From Levy and Naeser, 1991.

several additional dates near 8.5 Ma. The two sigma interval attached to the youngest age, of 4.8 Ma, is 2.5. Thus, there is very high confidence (over 90%) that the age of heating of this crystal was between 2.3 Ma and 7.3 Ma, with the highest probability for a specific age being 4.8 Ma. The same interpretation of confidence intervals applies to the other ages given. Clearly, characterizing these age data as being within the age range 10-12 Ma, given "analytical uncertainty," is incorrect. It is on this inaccurate basis that the Panel states that (p. 3):

"The preponderance of features ascribed to ascending water clearly (1) were related to the much older (13-10 million years old (Ma)) volcanic eruptive process that produced the rocks (ash-flow tufts) in which the features appear,..."

This conclusion is actually directly contradicted by the age data cited.

This issue is extremely important in that these are the only age data used in the NAS report to substantiate the claim that the last and final hydrothermal event occurred some 13 to 10 Ma ago. Age data from uranium series dating of calcites from veins at depth as well as potassium-argon dates from zeolites, which are commonly produced by hydrothermal alteration of volcanic glasses, were ignored by the Panel. However, as shown in Figure 2, many young ages are present in these data as well, some as young as 30 ka. In view of the preceding description of what is actually represented in the zircon age data, and in view of the zeolite and calcite vein age data, it is evident that high temperature annealing of fission tracks occurred at times much more recently than 10 Ma and that related hydrothermal alteration produced the observed young zeolites along with the recent calcite and opal veins throughout the mountain. Indeed, it is likely that analysis of additional zircon samples would show more recent ages, like the age data from the zeolites and calcites. Therefore, contrary to the Panel's statements, the age data actually support the occurrence of recent (post-Timber Mountain) hydrothermal activity rather than providing evidence against it.

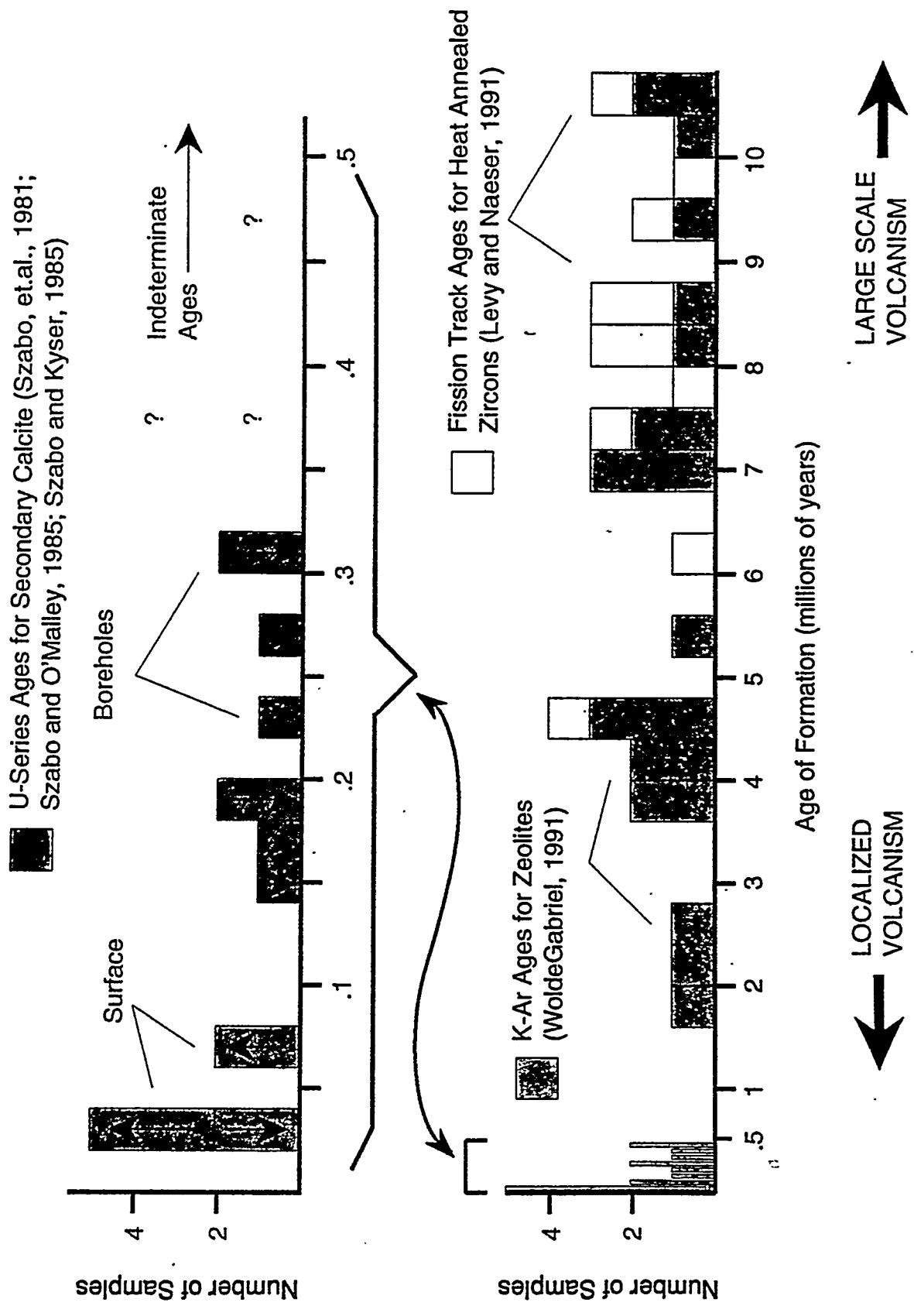


Figure 2. Ages of Fluid Alterations at Yucca Mountain

Field Observations: Spring Mounds, Faults and Surface Calcretes, Zeolites and Glass

Besides these misleading characterizations of important age data, the Panel has also characterized field observations inaccurately. One example is their statement that the Quaternary hydrothermal spring closest to Yucca Mountain is at Travertine Point, some 55 km away (p. 130). This statement is not correct: the hot springs at Oasis Valley just north of Beatty, Nevada, which were visited by the Panel, are only 25 km from the site. Further, they use the Travertine Point mound deposits to make the argument that springs at Yucca Mountain would also have to produce mounds, implying that all springs should produce mounds regardless of their topographic location or the chemical content of the water. However, the nearby springs at Oasis Valley do not now appear to be forming mounds. Likewise other springs in the region, at Boulder Dam and Dixie Valley, are not producing mounds. On the other hand, some of the many hot springs at Tecopa, CA (which is in the general area) are producing mounds, but others in this same area are not.

Consequently, the Panel has generalized from one example to establish a necessary criterion for ancient spring activity (the presence of mounds) and apparently presumed that the near proximity of the example to Yucca Mountain would provide the necessary justification. However, they are wrong on all counts: the example used is not the closest to Yucca Mountain, and mounds are sufficient but not necessary to establish spring activity. Indeed, water emerging from fault zones on a steep slope would not be expected to produce mineral mounds, but instead should produce slope parallel deposits, such as the calcrete deposits at Trench 14 and around Busted Butte.

Yet another example of importance is the Panels' statement (p. 33) in response to the idea that the observed calcretes at Busted Butte are produced by water flowing from up-slope fault zones. Here the Panel report rejects the idea on the basis of their own observation that there are no faults up-slope from these deposits. However, available geologic maps show at least one major fault zone at higher elevations at Busted Butte,

contrary to this statement.

These two examples are important in that the Panel uses lines of argument built upon these statements to assert, in their overall conclusion statement, that:

"The preponderance of features ascribed to ascending water clearly... (2) contained contradictions or inconsistencies that made an upwelling ground - water origin geologically impossible or unreasonable,..."

Another line of "evidence," considered by the Panel as contradictory or inconsistent with an upwelling water origin, is the zeolite and glass distribution with depth. Specifically citing the depth distribution of zeolites and glass as its evidence, the Panel states (p. 48):

"The boundary between the altered and vitric tuffs indicated that the water reached its highest levels and receded downward from 12.8-11.6 Ma, and that since that time the water level at central Yucca Mountain has probably not risen more than 60 m above its present position."

However, it is not possible to find the support cited for this conclusion from the actual data, which are shown in Figure 3. In particular, the observations show that, in some drill holes, glass is present hundreds of meters below the present water table. Further, zeolites are also present hundreds of meters above the water table. Thus, the distributions of zeolite and glass do not produce a simple relationship with the water table, that is both glass and zeolite occur above and below the water table making it impossible to establish a boundary and an ancient receding level for the water table based on these data.

In regard to the latter, it is important to point out that the Panel did not mention that the K-Ar dates of the zeolites in question range from 2 to about 10 Ma, as shown in Figure 2, and are much younger than the host ignimbrites. Further, the youngest zeolites are near the surface and the oldest are at depths below the water table. If the water table reached its highest level at 12.8 - 11.6 Ma and receded downward from that

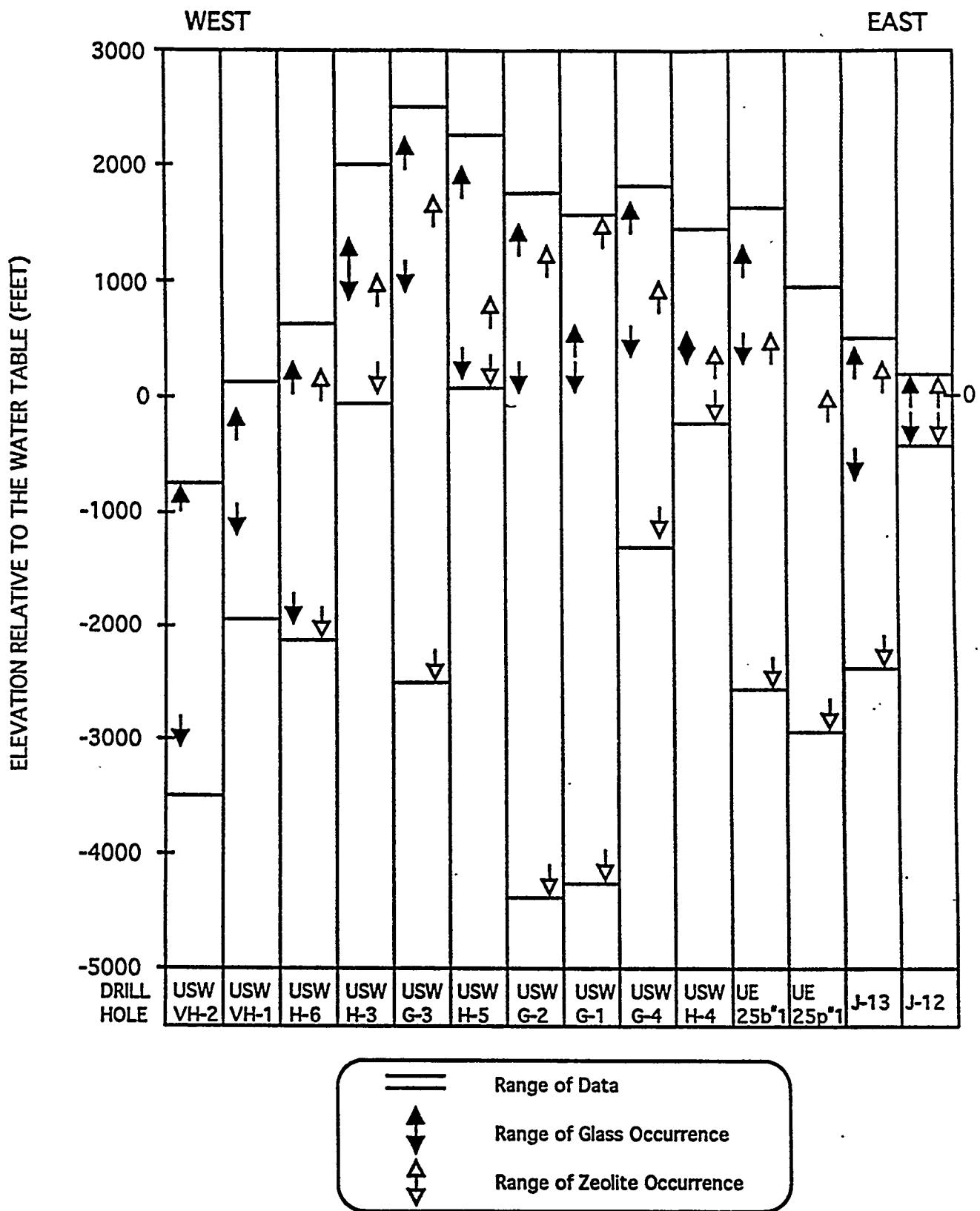


Figure 3. Distributions of glass and zeolite relative to the water table. Drill hole locations are distributed within and very near the proposed repository at Yucca Mountain with the exceptions of VH-2 and VH-1 in Crater Flat to the West and J-13 and J-12 in Jackass Flat to the East.

time to its present level, the opposite depth-age relationship for the post-10 Ma zeolites would be expected. Indeed, this depth-age relationship is what would be expected for an upwelling hydrothermal origin of the zeolites. Furthermore, this is the process generally accepted as being responsible for zeolitization in any case.

Isotopic Data: Comparisons Between Vein Calcites and Ground Water

A second major problem with the Panel report is that the strong conclusions produced by the Panel are either not reasonably supported by the evidence presented or are inconsistent with data and analysis results not cited in the report. This represents a class of problems differing from the previous cases, where the data cited are at least consistent with what is reported in the literature (though insufficient to support the conclusions drawn). However, the data cited are, nevertheless, not sufficient to support the conclusions drawn.

An example of this situation arises from the Panel's statements (e.g., p. 52 & p. 148) that the isotopic ratios for strontium, uranium and thorium for the near-surface vein calcites at Trench 14 and Busted Butte do not match the measured ground water values and therefore that ground water cannot have been responsible for their deposition. Here they compare the isotopic ratios in the calcites to those characteristic of meteoric water at shallow depths below the water table level. At these depths the water resides in volcanic tuffs and does indeed have discordant isotope ratios relative to the surface calcites. However, what the Panel fails to mention is that the isotopic characteristics of the water change with depth, since its isotopic character depends on the host rock properties. Specifically, a strontium isotope ratio measurement from the only well that penetrated the Paleozoic limestones at Yucca Mountain gives a value significantly higher than those from the shallower water in the tuffs, and close to the moderately high values observed in the surface veins in question. Further, while values from yet deeper water, including that in the Precambrian below the limestones, have not yet been

obtained at the site, the samples from older rocks at other sites, particularly in Precambrian rocks and Paleozoic shales, show very high strontium isotopic ratios in the range and higher than those observed in the Yucca Mountain and Busted Butte calcite veins, which average around .7125. The relationships of strontium ratios to rock types are illustrated by the data compiled in Table 1, where rhyolites and tuffs have low ratios around .707, limestones have ratios near .709 while Precambrian rocks have high ratios near .717.

Consequently, it is very likely that if water were convected upward from depths of the order of 3 km or deeper at Yucca Mountain it would have high strontium isotopic ratios and when mixed with the shallower water, which has lower strontium ratios, would produce the moderately high strontium isotopic ratio values observed in the near surface vein calcites. A similar argument applies to the other isotopes, although in the case of uranium series isotopes it is more complex (Archambeau and Price, 1991).

It is significant that the Panel offered no discussion of why the strontium ratios at Trench 14 and elsewhere at Yucca Mountain are so high, relative to observed limestone values. Certainly if these vein and associated calcrete deposits are simply due to the evaporation of rainwater carrying calcium and strontium picked up in solution from wind blown dust from (rather distant) limestone outcrops, as is asserted by the Panel, then one would expect to see strontium ratios near the limestone values of .709 rather than the much higher values that average .7125. Surely one could make the argument that there is no apparent support for such a pedogenic origin based on the isotopic data. Indeed there is every reason to doubt this hypothesis in view of the very discordant values observed in the strontium ratios of the surface calcites at Yucca Mountain relative to the values to be expected from the available sources of wind-transported calcite near Yucca Mountain.

Thus, the Panel has ignored important consequences of a "pedogenic origin" for the calcites and have also ignored the possibility of upwelling from greater crustal depths, where it is known that the isotopic ratios of the water would be different from those

Location	Rock	$^{87}\text{Sr}/^{86}\text{Sr}$	Source	Note
Unaltered Ignimbrites				
Long Valley Caldera	Inyo Domes Rhyolites	0.70630	Goff et al. (1990)	mean of 3 samples
do	do	0.70606	do	mean of 7 samples
do	Mafic and Intermediate	0.70630	do	mean of 3 samples
do	Moat Rhyolites	0.70601	do	mean of 6 samples
do	Early Rhyolites	0.70665	do	mean of 2 samples
do	do	0.70716	do	hydrothermally alt
do	do	0.70742	do	do
do	Bishop Tuff	0.7070	do	mean of 2 samples
do	do	0.70713	do	mean of 6 samples
do	do	0.70645	do	sanidine separates
do	do	0.70745	do	hydrothermally alt
do	Pre-caldera Volcanic	0.70610	do	mean of 3 samples
representative mean value: 0.70667				
Paleozoic Carbonates				
Spring Mountains	Limestone	0.70913	Peterman (1990)	outcrop
do	do	0.70823	do	do
do	do	0.70837	do	do
Ash Meadows	do	0.70990	do	do
Rock Valley	do	0.70934	do	do
representative mean value: 0.70899				
The Precambrian Basement				
Round Vly. Peak, CA	Schist	0.71656	Goff et al. (1990)	PC-derivative
do	Hornfels	0.72201	do	do
do	Sandstone	0.71126	do	do
Dish Hill, CA	Granodiorite	0.7177	Peterman et al (1970)	xenolith
representative mean value: 0.71688				

Table 1. Strontium isotopic ratios of unaltered ignimbrites, paleozoic carbonates and Precambrian rocks of the western Basin and Range Province. The high strontium isotopic ratio (> 0.71) of Yucca Mountain alteration products and calcite veins is indicative of a deep crustal source.

in the shallow water. Further, it is known, or can be inferred, that the ratios from the deep sources of water would be close to those observed in the vein calcites. Instead, they have implicitly assumed that either convection from such large depths does not happen or simply ignored the evidence of the changing isotopic character of the water with depth and formed the conclusion that ground water in general cannot be responsible for the calcite vein deposits at the site. Since Wood and King (1992) show that the volumes of outflow at the surface (approximately .5 km³) in the vicinity of the Borah Peak (Idaho) and Hebgen Lake (Montana) earthquakes can be explained as upward water flow ("seismic pumping") along fracture zones from depths at least as great as 5 km, it is clear that the possibility of upwelling of water from the Paleozoic and Precambrian should have been addressed by the Panel. Since they neither take note of the upwelling evidence given by Wood and King nor consider the changing isotopic ratios in the water with depth, their conclusion appears inappropriate and, in fact, might clearly be reversed when all the pertinent data are considered.

Indeed, even the limited data used by the Panel to support their conclusions can be interpreted quite differently. Specifically, the shallow water near the top of the water table should be representative of infiltrating rain water in areas at and near Yucca Mountain where there is no upwelling of convected water from depth. Such "sink areas" are extensive at Yucca Mountain and the water at depth should be representative of infiltrating rain water. If this water does not have isotopic characteristics matching the vein calcites, which it does not since the strontium ratio for such water is .7105, then the logical conclusion is that infiltrating meteoric water (which would have taken any available calcium and strontium from wind-blown dust into solution) does not have isotopic characteristics that are compatible with the observed vein calcites. This observation, as well as those given previously, contradict the Panel's general conclusion that these vein calcites are "*classic examples of arid soil characteristics recognized world-wide.*" Further, rather than showing that the isotopic character of the vein minerals versus that of the shallow ground water rules out upwelling ground water as a source of the calcite-opal veins observed, the lack of agreement between the isotopic

characteristics of the vein calcites and the shallow water at Yucca Mountain can be interpreted to mean that pedogenic hypothesis advanced is not supported by the pertinent isotopic data.

Water Level Changes at Devils Hole

Another example of a conclusion that is not reasonably supported by the evidence and data cited is the water level data at nearby Devils Hole. The Panel cites evidence (pp. 35, 55) that the ground water level exposed in the open cavern at this location has not fluctuated by more than 10 meters in the last 45 ka. In addition the Panel cites evidence from other studies that imply that the water level has been below the land level, which is 16 meters above the ground water level, for the last several hundred thousand years. However, the Panel fails to mention, or take account of the fact, that the Devils Hole Cavern occurs in an isolated outcrop with its opening elevated above the surrounding area and that within this nearby area there are many active springs. Thus, any rise in the water table would result in greater surface outflow from the active springs and so prevent any rise in the Devils Hole water level above about 10 meters. Consequently, the water level data in the Devils Hole Cavern does not reflect upward rises in the water table, although declines in the level should be correlated with declines in the water table in the area. In this regard, there is some evidence that the water level in the cavern may have been lower in the past than at present. In any case however, the Panel's argument that the water table has probably been stable for a long period of time, based on lack of evidence for any rise in the water level at Devils Hole greater than 10 meters, is not correct.

Age Data, Low Grade Metamorphic Alteration and Temperature Data

The final area of major concern with the Panel's report is the neglect of the very large body of data relating to the ages and character of hydrothermal alterations at the

site. The Panel uses very limited data, and principally the zircon age data previously discussed, to argue that the last hydrothermal event occurred about 10-12 Ma ago. However, in addition to the zircon age data, which actually implies much more recent activity, there is an additional body of data that also indicates that there has been on-going hydrothermal activity.

This data involves the age data shown in Figure 2 in combination with paleogeotherm estimates inferred from oxygen isotopes, rock alteration temperatures from zeolitization and illitization processes in rocks at Yucca Mountain, vein formation temperatures from fluid inclusions, and finally, zircon annealing temperatures from the samples at Trench 14 and Busted Butte. All of this inferred temperature data, shown in Figure 4, indicate high temperatures and high geothermal gradients existent at Yucca Mountain in the past. Since the age data shown in Figure 2 are from samples in close proximity to the locations sampled for the temperature estimates, and in the case of the zircons are the same samples used to estimate annealing temperatures, there is little doubt that the high temperatures and gradients are associated with very recent hydrothermal activity at Yucca Mountain. In particular, the K-Ar and uranium-series dates for zeolites and calcium carbonate vein material, respectively, indicate episodic and moderate to high temperature hydrothermal activity that has continued from 13 Ma to essentially the present. In addition, the zircon ages and annealing temperatures also indicate post-Timber Mountain hydrothermal activity involving quite high temperatures for the fluids involved. Finally, all the geothermal gradients inferred from heat flow and oxygen isotope data are sufficient to produce convection and are therefore consistent with a history of hydrothermal activity.

The fact that the Panel did not consider any of the data pertaining to paleo-temperatures and ignored all the age data, except that for the zircon ages which they misrepresented, has resulted in a description of the recent geologic and hydrologic history of the site that is almost certainly incorrect. Indeed, the only uncertainty that might still be entertained is whether the youngest ages, of less than 500 ka, are correlated with the high temperatures indicated in Figure 4. This can be cleared up

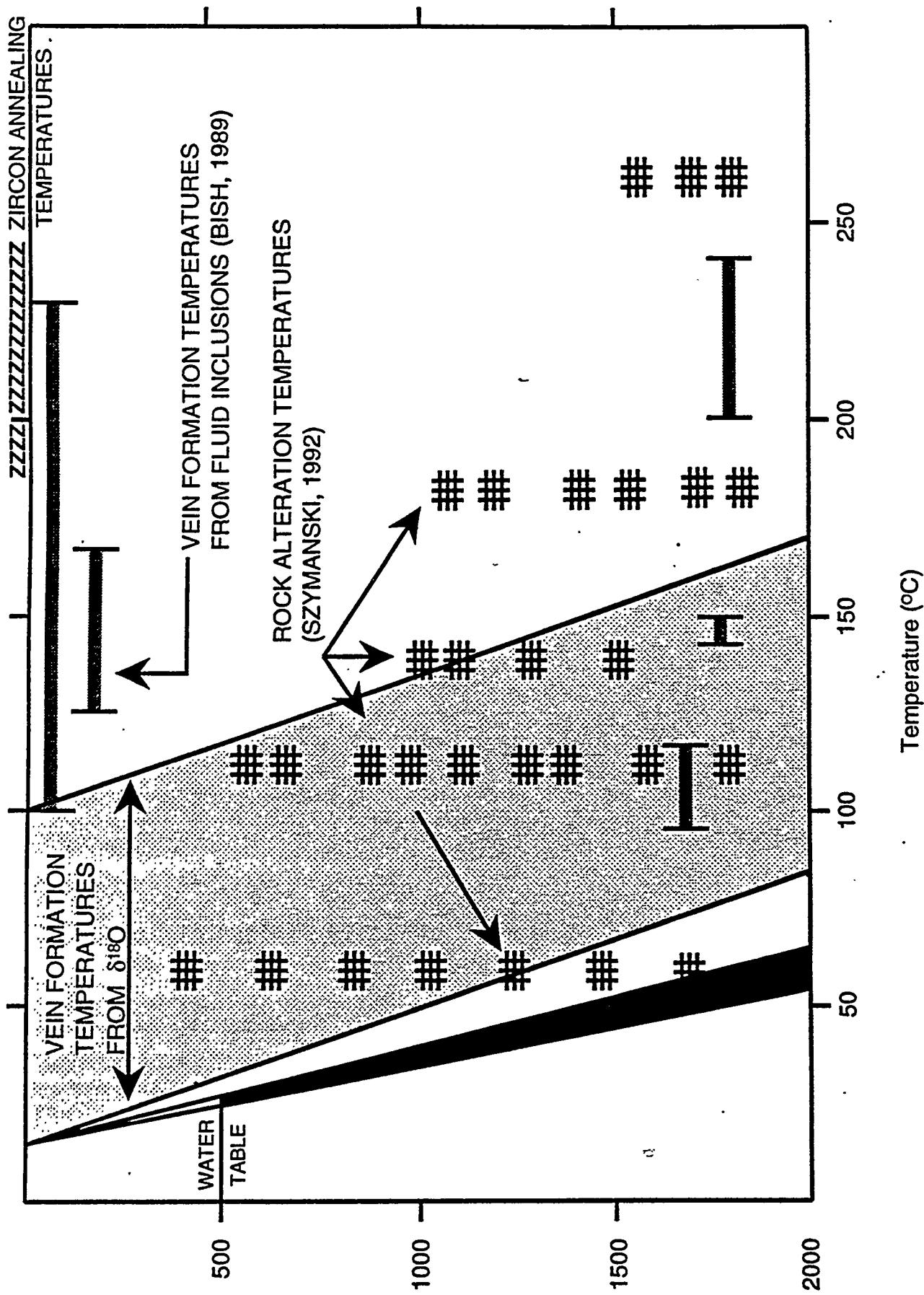


Figure 4. Borehole samples from Yucca Mountain reveal alteration products formed at high temperatures, indicating that the site has been invaded by high temperature fluids.

by additional sampling of course, but in any case there is no reasonable doubt that hydrothermal alteration and deposition occurred well after the time of 10 to 12 million years ago claimed by the Panel. Once this Panel conclusion is recognized as unsupportable in the face of the available quantitative age and paleo-temperature data, it only becomes a question of how frequently and how recently the episodic hydrothermal activity has occurred. The available data shown in Figures 2 and 4 clearly suggest that it has been frequent enough and recent enough to justify the belief that it will most likely continue and that it could occur at any time in the future.

In addition to ignoring age and paleo-temperature data, the Panel did not address the significance of the reported mineral enrichment of interstitial fluids extracted from pores within the tuffs above the water table (Smith, 1991). Relative to local fluids within fractures in the tuffs, the interstitial fluids are strongly enriched not only in alkali-earth elements, but also in transition, base and noble metals and rare earth elements (REE) which at least suggest, if not require, a hydrothermal origin. Table 2 indicates the observed enrichment of several elements found in this trapped water, expressed as a ratio of abundances relative to the element content in nearby well water. Clearly, the presence of noble and base metals is indicative of a hydrothermal fluid. Further, in addition to an overall enrichment of REE, there is an unusual enrichment of heavy REE relative to light REE that is not shared by the host ignimbrites. This enrichment is illustrated in Figure 5 where the normalized REE abundances versus increasing REE atomic weight are shown for the interstitial fluids (a) and local ignimbrites (b). Clearly the abundance trend versus atomic weight is quite different for the ignimbrites compared to the interstitial water. Specifically, the relative enrichment of heavy REE in the interstitial water is conspicuous and since it is also observed elsewhere for hydrothermal solutions that are concentrated in CO_2 (Michard and Albarede, 1986; Michard et al., 1987), it is certainly likely that these fluids are remnants of late hydrothermal fluids.

Table 2
Mineral Enrichment of Vadose-Zone Interstitial Fluids

ELEMENT	ENRICHMENT Ratio *
Magnesium	10
Calcium	8
Nickel	1000
Copper	50
Zinc	45
Rubidium	2
Strontium	30
Yttrium	100
Molybdenum	300
Iodine	20
Tungsten	300
Platinum	**
Gold	**
Titanium	20

*Data are from borehole UZ#4 (interstitial fluids) normalized by J-12 and J-13 well waters (Smith, 1991).

**Well waters contained no measured gold and platinum. Interstitial fluids contained .2 ppb for both metals.

Table 2. Mineral enrichment of vadose-zone interstitial fluids relative to well waters residing in ignimbrite fractures.

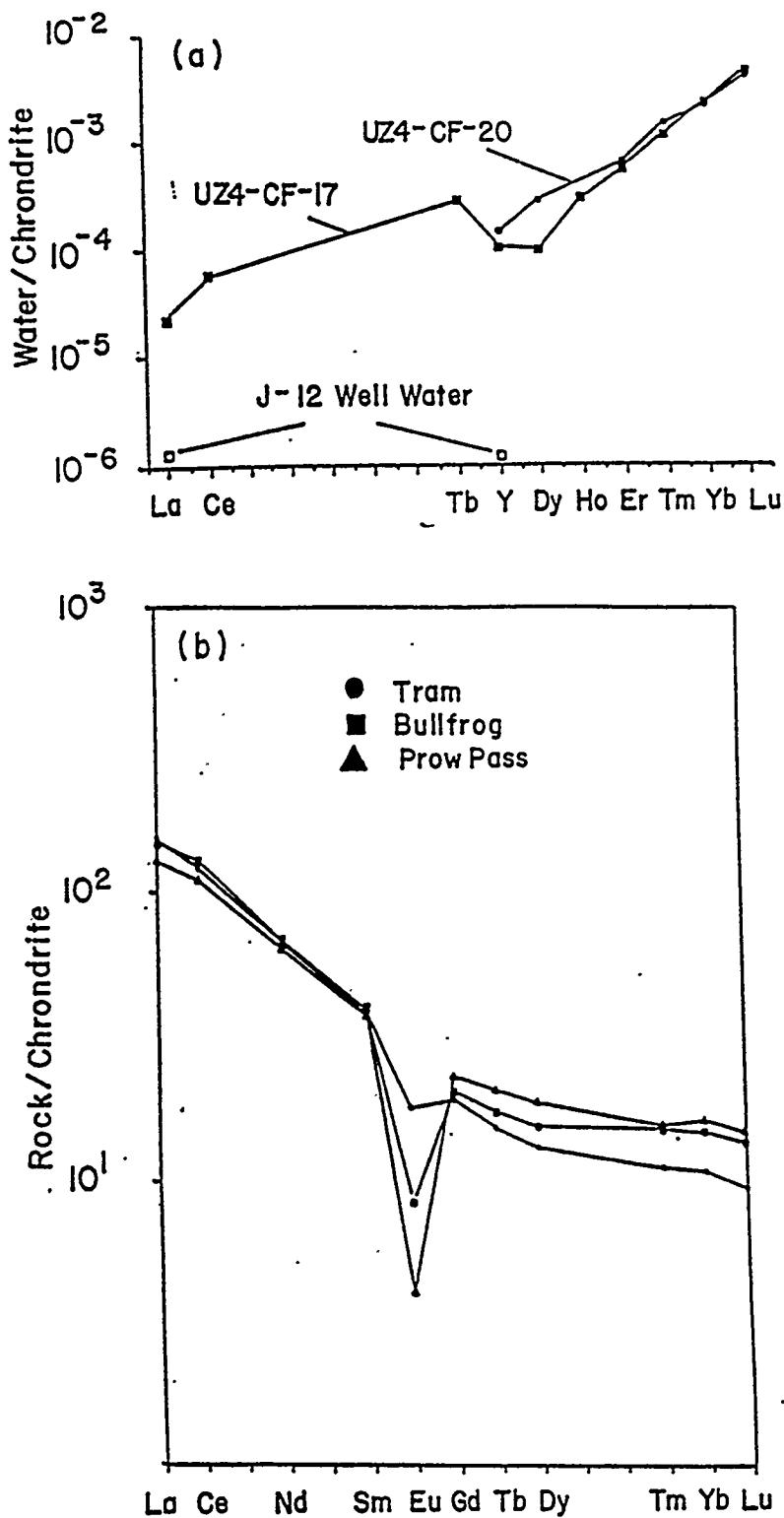


Figure 5. Chondrite-normalized REE abundance patterns. a.) interstitial fluids and well water residing in ignimbrite fractures: data from Smith (1991). b.) Crater Flat ignimbrites: data from Scott and Castellanos (1984). Heavy REE enrichment for interstitial fluids is due to high CO_2 pressure.

The inference of a high CO₂ content for these remnant hydrothermal fluids is important in that a high gas content would be consistent with an interpretation of gas assisted fragmentation and brecciation during hydrothermal fluid intrusion and account for observed intense brecciations of the country rock associated with late carbonatization at many sites at Yucca Mountain. This inference, while not conclusive in itself, does certainly bring into question the Panel's conclusion that (p. 46):

"...there is no need for, or good evidence in support of, upwelling of deep hot waters to account for the brecciation (of near-surface country rocks) or silica - carbonate cementation."

If the Panel had presented the fluid inclusion data along with the temperature and age data in their report, it seems unlikely that they could have made such a statement or, if made, have made it sound plausible in the face of the evidence.

A related Panel statement involves the fault breccia cement at Trench 14. The Panel conclusion states (p. 44):

"...that the fault breccia cement at Trench 14 and Busted Butte is of pedogenic or surficial origin, based on the presence of older detrital zircons, grain size and structure characteristics, and is not of hydrothermal origin."

As noted earlier, the zircons are not as old as indicated by the Panel and in any case do not provide an age estimate for low to moderate temperature hydrothermal deposition (see the temperature range for zircon fission track annealing indicated in Figure 4), while the small grain size of the calcite cement could be expected to occur as a consequence of rapid release of CO₂ from a hydrothermal fluid near or at the surface (Archambeau and Price, 1991). Further, the "structure characteristics" referred to by the Panel are precisely those interpreted by others, such as Hansen et al. (1987), as being characteristic of hydrothermal brecciation.

Thus, the strong conclusion drawn by the Panel is certainly not warranted by the observations they cite, in that other interpretations are at least as plausible if not preferable. But beyond these alternative interpretations, it is once again evident that the

Panel should have used additional available data to infer the origins of the silica-carbonate breccia cements and veins at Yucca Mountain. In this regard Table 3 provides a clear indication of the unusual enrichment of the breccia cement in base and noble metals relative to the stratigraphically equivalent background values for the tuffs at Trench 14. The results in the third column are the median values for 25 analyses of nine breccia samples while the fourth column indicates the significant enrichment of the most strongly mineralized specimen. The fifth column shows that the degree of enrichment of the interstitial fluids (discussed earlier) is comparable with that of the more strongly mineralized breccia samples. Such enrichment contradicts the hypothesis of a pedogenic origin for the breccia cements and combined with the previously mentioned age and temperature data is strong evidence for a hydrothermal origin of the breccia, which is of post-Timber Mountain age.

Beyond the omissions of the data and results already mentioned, the Panel does not address several other topics and related data of considerable importance. In this regard, in situ stress measurements, such as those by Healy et. al. (1984) and Stock et. al. (1984, 1986), are clearly critical to an assessment of geodynamic stability of the site. These observations were not considered by the Panel. However, contrary to the Panel's assessment that the Yucca Mountain area is not likely to experience a large earthquake in the near future, the results from Healy et al. and Stock et. al. imply the opposite. Indeed, the recent 5.6 magnitude earthquake at Little Skull Mountain, 15 km southeast of Yucca Mountain, also indicates that an unstable stress state, rather than a quasi-stable state, actually prevails.

Consequently, at least in part because of their lack of consideration of a large body of the most quantitative and unequivocal data, the Panel reached many conclusions that are not supported by the complete body of data that exists.

Table 3
Mineral Enrichment of Breccia Cement

ELEMENT	ENRICHMENT			
	TIVA CANYON LITHOPHYSAL TUFF FROM EXILE HILL *	MEDIAN, TRENCH #14 BRECCIA CEMENT *	MAXIMUM, TRENCH #14 BRECCIA CEMENT *	INTERSTITIAL FLUIDS **
Ag	2	2	16	
As	1	3.6	36	
Au	<1	2	5	
Cu	.25	1	4	50
Mo	7	18	650	300
Pb	14	65	610	1-5
Sb	<1	25	100	
Zn	44	90	33	45
Bi	<1	<1	<1	

*Data from Weiss (1990); the maximum values of enrichment are for a single sample (3SW195B) with the highest overall mineral enrichment relative to average concentrations for the Yucca Mountain area (Castor et al., 1989).

**Data from Smith (1991); enrichment relative to well water.

Table 3. Mineral enrichment of breccia cement: results for lithophysal tuff and interstitial fluids are shown for comparison.

General Comments on the Panel Report

In addition to a general disregard of important quantitative data and a rather cavalier approach to elementary logic, the Panel not only distorted some of the data and interpretations reported in the literature (such as the zircon age data) but also misrepresented the concepts described by Szymanski in his 1989 report on hydro-tectonic activity at the Yucca Mountain site. To make matters worse, the Panel also misrepresented the information given to them during a presentation by the minority members of the DOE External Review Panel (Archambeau and Price). Specifically, the NAS/NRC Panel states, on page 129 of their report:

"It should be noted that the charge to the panel included an evaluation of the particular concepts described in the report by Szymanski (1989). Those concepts involved seismic pumping as the primary mechanism for driving the deep ground water to the surface in a cyclic progression of crustal stress changes. The panel evaluated the geologic evidence presented for this process and found both the evidence and the seismic pumping model inadequate to support the consequences attributed to them. As the panel was concluding its studies, the "minority" members of the 5 member external review panel selected by DOE and Szymanski to review his report informed the NAS panel that both the interpretation of some of the evidence and the model itself had changed: that Szymanski no longer believed that seismic pumping alone could drive the water up as high as he had stated in his report, and that he now had a new concept involving a thermally driven hydro-tectonic cycle. This information was presented at the NAS panel's last meeting. Although there was no time left for the NAS panel to give consideration to a new thesis, nor was there a written document that could be evaluated, the cyclical concept as presented to the NAS panel appeared to have little validity, given that the panel is convinced that the geologic evidence refutes the assertion that ground water has risen repeatedly 100 meters or more in the recent geologic past. Because an essential part of the "cycle" has not yet happened, there is no basis for postulating a cyclical process whatever the proposed mechanisms involved."

In referring to the minority members' report, the Panel alleges that they were informed that "both the interpretation of some of the evidence and the model itself had changed" and then go on to elaborate that Szymanski now "had a new concept involving a thermally driven hydro-tectonic cycle." Both of these statements are false.

Specifically, these statements were not made by the minority members. Indeed the material distributed to the NAS Panel by the minority members describes, in very specific terms, the full concept advanced by Szymanski in his 1989 report which includes the concept of a hydrotectonic cycle involving *both* seismic pumping and thermally driven convection of the ground water following a tectonic event, such as an earthquake. This combined response to changes in the hydrologic system was considered to be the cause of upwelling water and associated mineral deposition at Yucca Mountain. Only if the minority members had contradicted their own written summary of Szymanski's 1989 report could they have made the statements attributed to them and that is simply not what occurred, nor realistically is it credible. Furthermore, the minority members presented a summary of their report to the NAS Panel in May of 1991 and submitted their complete report to the DOE in November of 1991. This final report reproduces the material made available to the NAS Panel. Therefore, it is a matter of record that the Panel had ample time to refer to the relevant material, long before they submitted their report in July of 1992, and in addition shows that they misquoted the minority members.

Beyond this distortion of the facts, the Panel misrepresented the content of Szymanski's 1989 report since they assert that he had changed his original concept of seismic pumping as the primary cause of water level changes and introduced a new concept involving thermally driven processes at a time well after writing his report. If the Panel had actually read Szymanski's report they would have found that this latter concept is discussed in considerable detail and was thought to be the principal mechanism for deposition of calcite throughout the mountain.

Therefore, one can only conclude that the Panel did not actually read Szymanski's report, or if they did read it they chose to misrepresent it. In either case this is hardly what would be expected from a NAS panel that is charged with the responsibility of evaluating a report. On this basis alone there would be reasonable grounds to seriously question the Panel's findings as it suggests an inclination to distort and misrepresent the record.

References

1. Archambeau, C. B., and M. J. Price, 1991. **An Assessment of J. S. Szymanski's Conceptual Hydro-Tectonic Model and Its Relevance to Hydrologic and Geologic Processes at the Proposed Yucca Mountain Nuclear Waste Repository.** Minority Report of the Special DOE Review Panel. U. S. Department of Energy, Las Vegas, Nevada.
2. Bish, D. L., and S. J. Chipera, 1989. **Revised Mineralogic Summary of Yucca Mountain, Nevada.** LANL Report, LA-11497-MS.
3. Castor, S. B., S. C. Feldman, and J. V. Tingley, 1989. **Mineral Evolution of the Yucca Mountain Addition, Nye County, Nevada.** Report to Science Applications International Corporation.
4. Hanson, G. N., V. R. Baker, P. M. Bethke, P. J. Hudleston, and G. R. Roquemore, 1987. **Report of the Peer Review Panel on the Proposed Program of Studies of the Calcite and Opaline-Silica Deposits in the Yucca Mountain Area, Nevada.** U. S. Department of Energy, Las Vegas, Nevada.
5. Healy, J. H., S. H. Hickman, M. D. Zoback, and W. L. Ellis, 1984. **Report on Televiewer Log and Stress Measurements in Core Hole USW-G1, Nevada Test Site, December 13-22, 1981.** USGS Open-File Report 84-15, 51 pp.
6. Levy, S., and C. Naeser, 1991. **Bedrock Breccias Along Fault Zone Near Yucca Mountain, Nevada.** Draft Los Alamos-USGS Paper.
7. Michard, A., and F. Albarede, 1986. **The REE Content of Some Hydrothermal Fluids.** Chemical Geology, p 55. Elsevier Science Publishers.
8. Michard, A., C. Beaucaire, and D. Michard, 1987. **Uranium and Rare Earth Elements in CO₂-Rich Waters From Vals-Les-Bains (France).** Geochimica et Cosmochimica Acta, 51.
9. Scott, R. B., and M. Castellanos, 1984. **Stratigraphic and Structural Relations of Volcanic Rocks in Drill Holes USW GU-3 and USW G-3, Yucca Mountain, Nye County, Nevada.** USGS Open-File Report 84-491, Denver, CO.

10. Smith, M. R., 1991. **Natural Radionuclide/Trace Element Hydrochemistry-Characterization of the Yucca Mountain Saturated (well waters) and Unsaturated (pore waters) Zone Ground Waters.** Pacific Northwest Laboratory, Richland, Washington.
11. Stock, J. M., J. H. Healy, J. Szitek, and L. Mastin, 1986. **Report on Televiewer Log and Stress Measurements in Holes USW G-3 and UE-25P#1, Yucca Mountain, Nevada Test Site.** USGS Open-File Report 86-369, 91 pp.
12. Stock, J. M., J. H. Healy, and S. H. Hickman, 1984. **Report on Televiewer Log and Stress Measurements in Core Hole USW-G2, Nevada Test Site.** USGS Open-File Report 84-172, 48 pp.
13. Szabo, B. J., W. J. Carr, and W. S. Gottschall, 1981. **Uranium-Thorium Dating of Quaternary Carbonate Accumulations in the Nevada Test Site Region, Southern Nevada.** USGS Open-File Report 81-119, Denver, CO.
14. Szabo, B. J., and P. A. O'Malley, 1985. **Uranium-Series Dating of Secondary Carbonate and Silica Precipitates Relating to Fault Movement in the Nevada Test Site Region and Caliche and Travertine Samples From the Amargosa Desert.** USGS Open-File Report 85-47, Denver, CO.
15. Szabo, B. J., and T. K. Kyser, 1990. **Ages and Stable-Isotopic Compositions of Secondary Calcite and Opal in Drill Cores From Tertiary Volcanic Rocks of the Yucca Mountain Area, Nevada.** Geological Society of America Bulletin, v. 102.
16. Szymanski, J. S., 1989. **Conceptual Considerations of the Yucca Mountain Groundwater System With Special Emphasis on the Adequacy of this System to Accommodate a High-Level Nuclear Waste Repository.** DOE Internal Report, U. S. Department of Energy, Las Vegas, Nevada.
17. Szymanski, J. S., 1992. **The Origin and History of Alteration and Carbonatization of the Yucca Mountain Ignimbrites.** DOE Internal Report, U. S. Department of Energy, Las Vegas, Nevada.
18. Weiss, S. I., 1990. **Yucca Mountain Project Monthly Report Task 3-August, 1990.** The State of Nevada Nuclear Waste Project Office, Agency for Nuclear Projects.
19. Weiss, S. I., D. C. Noble, and L. T. Larson, 1990. **Task 3: Evaluation of Mineral Resource Potential, Caldera Geology and Volcano-Tectonic Framework at and Near Yucca Mountain.** Report for October 1989 - September, 1990. Department of Geological Sciences, Mackay School of Mines, University of Nevada-Reno.

20. WoldeGabriel, G., 1990. **Diagenetic Minerals, D/Ar Data, and Alteration History in the Yucca Mountain, Nevada: A Candidate High-Level Radioactive Waste Repository.** Los Alamos National Laboratory draft paper.
21. Wood, R. M., and G. C. P. King, 1992. **Hydrological Signatures of Earthquake Strain.** Submitted to Journal of Geophysical Research.